

COMPARATIVE ANALYSIS OF THE ENVIRONMENTAL IMPACT OF ALTERNATIVE TRANSPORTATION FUELS

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INTRODUCTION

Emissions from motor vehicles contribute significantly to air pollution problems. Despite new emission standards and advances in motor vehicle emission control technology, many areas in the country are still projected to have air pollution problems in the year 2000 and beyond. It appears unlikely that there will continue to be significant declines in emissions from gasoline and diesel-powered vehicles. This situation has spurred interest in alternative fuels for transportation.

Substituting alternative transportation fuels for gasoline and diesel fuel may improve air quality in the United States. The goal of this paper is to analyze the impact of alternative transportation fuels on attainment of The National Ambient Air Quality Standards for ozone and carbon monoxide (CO). Although cost and other consumer acceptance factors are not analyzed, all the alternative fuels studied are considered feasible for use by the general public. The term "alternative fuel" is used throughout this report to mean any non-gasoline or diesel fuel, including gasoline mixtures.

This report concentrates on light duty applications of alternative fuels, because light-duty vehicles play a much greater role in ozone and CO non-attainment than heavy-duty vehicles. The emphasis in this report is on how methanol, compressed natural gas (CNG), and liquified petroleum gas (LPG) compare with gasoline.

IMPACT OF ALTERNATIVE FUELS ON ATTAINMENT OF THE NAAQS FOR OZONE

One of the most persistent air quality problems in the U.S. has been the attainment of the NAAQS for ozone. Currently, approximately 90 million people live in areas that have one or more violations of the ambient ozone standard, and a downward trend in ozone levels is not evident. In 1988 there were more ozone violations than in many of the previous years.

Ozone is caused by atmospheric photochemical reactions involving volatile organic compounds (VOCs) and oxides of nitrogen (NO_x). Mobile sources account for about half of these emissions. The ozone formation rate is greater at higher ambient temperatures.

VOCs are emitted from mobile sources as either tailpipe or evaporative emissions. Tailpipe emissions occur as a result of incomplete combustion and/or chemical reactions during combustion. NO_x is largely produced by reactions between nitrogen and oxygen at high temperatures.

EPA policy emphasizes that states should control ozone by reducing VOCs rather than NO_x emissions. However, in areas dominated by VOC emissions (i.e., they have a high HC to NO_x ratio in the ambient air) there is some evidence that reducing NO_x emissions, as well as VOCs, helps reduce ozone.

Impact of Alternative Fuels on Reactive VOC Emissions

VOCs emitted from mobile sources typically are termed hydrocarbons (HCs). In 1988 most (92 percent) of the HC emissions from mobile sources were from light-duty gasoline-powered vehicles (1). Therefore, these vehicles are the target for additional HC controls.

An analysis of VOC emissions impacts must consider both exhaust and evaporative emissions. Furthermore, the photochemical reactivity of these emissions must be considered.

The State of California recently quantified the composite reactivity of emissions from vehicles powered by different fuels. In their study, they speciated emissions for different fuels and calculated the mass-weighted reactivity of the total vehicle emissions. The results of their study are summarized on Table 1 (1). Non-methane VOC emissions from natural gas-powered vehicles are less reactive than those from vehicles powered by other alternative fuels; they are less than half as reactive as non-methane hydrocarbons (NMHC) from gasoline-powered vehicles. Methane emissions were excluded because they have negligible reactivity. VOC emissions from methanol-fueled vehicles include methanol and are estimated to be between 50 and 56 percent as reactive as those from gasoline-powered vehicles. The lower percentage assumes low formaldehyde emission rates (15 mg/mile).

The following discussion compares non-methane VOC emission rates for vehicles powered by different fuels. Exhaust and evaporative emissions rates that are reported in publicly available sources are analyzed.

Reactive VOC Exhaust Emissions for Different Fuels - Reactive VOC emissions from alternative fueled vehicles include non-methane hydrocarbons (NMHCs), methanol, and formaldehyde. Figure 1 summarizes information on NMHC and formaldehyde exhaust emissions from light-duty vehicles burning different fuels. Although a range of values is shown on Figure 1, almost all the emission tests were performed on low-mileage vehicles, so the range may still underestimate in-use emissions. The M100 (100% methanol) numbers are for advanced, dedicated prototypes, while the CNG and LPG numbers are more representative of production dual-fuel vehicles.

For reference purposes, two estimates are presented for VOC exhaust emissions from gasoline-powered vehicles. The high estimate was generated by MOBILE4 (EPA's mobile source emission factor model) for a fleet composed almost entirely of 1981 and newer automobiles. The low estimate for gasoline-powered vehicles equals the exhaust emission standard for 1981 and newer vehicles.

Table 2 summarizes the range of methanol exhaust emissions in grams per mile that were found in the database for M85- and M100-fueled vehicles. Note that the high number (1.6 g/mi) for M85 (85% methanol, 15% gasoline) was a 50,000 mile projection made by EPA (2), so it most likely represents the in-use emission factor in grams per mile. The high value for M100 (1.7 g/mi) is the average of emission test results on vehicles operated on greater than 90 percent methanol (3). If in-use vehicles emit methanol at rates close to the high range shown on Table 2, some ozone impacts, in addition to those from NMHC and formaldehyde emissions, are likely from methanol-fueled vehicles.

Evaporative Emissions for Different Fuels - Evaporative emissions are composed of stationary evaporative losses (hotsoak and diurnal losses), running evaporative losses, and refueling losses. VOC emissions due to fuel evaporation will vary greatly for the different alternative fuels.

Figure 2 shows a comparison of stationary NMHC evaporative losses for different fuels. The database contains information on stationary evaporative losses for vehicles fueled with M85; MOBILE4 was used to estimate evaporative losses from gasoline-powered vehicles. The high value (0.14 g/mi) for M85 shown on Figure 2 is a 50,000 mile in-use projection made by EPA (4). The low value for M85 is based upon test results for two advanced prototype vehicles (5).

Few data are available on the amount of VOC's that is emitted due to running losses or refueling losses for the different alternative fuels. Table 3 shows estimated running and refueling NMHC losses based upon engineering judgment.

Table 4 summarizes the reported range of methanol evaporative emissions while the vehicle is stationary (hot soak and diurnal losses). The high value for M85 (0.37 g/mi) is the 50,000 in-use estimate by EPA (2). The low value (0.02 g/mi) is based on two advanced prototypes tested by the California Air Resource Board (4). Only one test result was available on advanced M100 vehicles (equivalent to 0.12 g/mi) (4).

Total Reactive VOC Emissions from Light-duty Vehicles for Different Fuels - Figure 3 shows estimates of the total exhaust and evaporative reactive VOC emissions from light-duty vehicles during periods when the ambient temperature ranges between 60° and 84°F. Methanol emissions are indicated by the shaded area. Because they have low NMHC emissions in the exhaust and negligible evaporative emission losses, dedicated CNG vehicles are estimated to emit the smallest amount of reactive VOC emissions.

The total emission values for M85 and M100 vehicles include methanol. The totals for M85 are similar to the MOBILE4 estimate for gasoline-powered vehicles, but as discussed earlier M85 vehicle emissions are less reactive than gasoline-powered vehicle emissions. Thus, there may be some ozone benefits for M85, but a clear benefit is not evident. M100-powered vehicles are estimated to have greater ozone benefits than M85 because they appear to emit much less NMHC. Dedicated LPG vehicles may have similar benefits to M100 vehicles; LPG may result in greater NMHC emissions than M100 but M100 will result in substantial methanol emissions.

Dual-fuel LPG and CNG vehicles will emit much greater amounts of VOCs than dedicated LPG and CNG vehicles because of evaporative NMHC losses.

NO_x Emissions for Different Alternative Fuels

The other precursor component in the atmospheric formation of ozone is oxides of nitrogen (NO_x). In 1988 about two-thirds of the mobile source NO_x emissions came from light-duty gasoline-powered vehicles.

Figure 4 compares estimated emissions from light-duty vehicles burning different alternative fuels with emissions from light-duty gasoline-powered vehicles. Unlike the case with NMHC emissions, MOBILE4 estimates of NO_x emissions from 1981 and newer light-duty vehicles are identical to the NO_x standard.

Considering that gasoline-powered vehicles can meet much more stringent emission levels than the Federal NO_x standard, none of the alternative fuels appears to offer clear advantages in reducing NO_x emissions from light-duty vehicles. Emission rates lower than the MOBILE4 estimates were observed for all the fuels; however, emission rates equal to or higher than the MOBILE4 estimates also were observed for most of the fuels. One can conclude that light-duty vehicles can be designed to burn alternative fuels such as CNG, LPG, or methanol and meet emission levels achievable by gasoline-powered vehicles; but it appears unlikely that large reductions are possible. Dual-fuel CNG vehicles are expected to emit about the same amount of NO_x as dedicated CNG vehicles with similar NO_x emission controls.

IMPACT OF ALTERNATIVE FUELS ON ATTAINMENT OF THE NAAQS FOR CARBON MONOXIDE

Exceedances of the NAAQS for carbon monoxide (CO) are less widespread than exceedances of the ozone standard. There has been a significant downward trend in ambient CO concentrations, but several tough CO attainment problems remain. Areas with extreme ambient conditions, such as Alaska and Colorado, are not projected to attain the CO standard without additional controls.

About 80 percent of the nationwide CO inventory is from mobile sources. And most (91 percent) of the mobile source CO emissions are from light-duty gasoline-powered vehicles (1).

Figure 5 shows the range of CO emissions that were observed for light-duty vehicles powered by different fuels. Two estimates of gasoline-powered CO emissions are provided as a reference. One is the MOBILE4 estimate for 1981 and newer vehicles, the other is the CO emission standard for 1981 and newer vehicles.

CNG-powered light-duty vehicles appear to have lower CO emissions than vehicles powered by other fuels. These levels were achieved by vehicles with both advanced emission controls and with no emission controls. These data indicate that CO emissions from CNG-powered vehicles are likely to be very low in actual use, because CO emission levels are less sensitive to vehicle technology or tampering. It is possible to run a CNG engine rich (too much fuel) which greatly increases CO emissions, but these cases should be identified in most inspection/maintenance programs or preventive maintenance checks.

LPG vehicle emissions are higher than CNG vehicle emissions, but are lower than the CO standard for 1981 and newer light-duty vehicles. When M85 and M100 vehicle emissions are compared with the CO emission standard, there appears to be no clear advantage for those fuels. The data are not adequate to project a CO emission value comparable to the MOBILE4 estimate for M85- and M100-powered vehicles. Because most of the emission tests were performed on low-mileage, well-maintained vehicles, it is likely that actual in-use emissions for those fuels would be much higher.

CONCLUSIONS

Impact of Alternative Fuels on Attainment of the NAAQS for Ozone - Efforts to attain the NAAQS for ozone would be enhanced if vehicle fleets in non-attainment areas consumed certain alternative fuels instead of gasoline. Dedicated CNG vehicles appear to have the greatest ozone benefits. LPG and M100 vehicles also offer significant ozone benefits. However, dual-fuel CNG or LPG vehicles and M85 vehicles (vehicles designed to burn mixtures of 85 percent methanol and 15 percent gasoline) may not be much better than gasoline vehicles. The primary reason for this is that evaporative volatile organic compounds (VOCs) emissions from storage of gasoline in the vehicle greatly increase their overall contribution to ozone formation.

This conclusion assumes that each fuel will displace a similar amount of gasoline. It does not consider consumer acceptance or infra-structure issues that will impact the market penetration of an alternative fuel.

Impact of Alternative Fuels on Attainment of the NAAQS for Carbon Monoxide (CO) - Both dedicated and dual-fuel CNG vehicles emit much less CO than gasoline-powered vehicles, so their use will help an area attain the CO NAAQS. LPG vehicles also appear to produce lower amount of CO than gasoline-powered vehicles, but they emit greater amounts than CNG-powered vehicles. Available data are not adequate to project the impact of methanol-fueled vehicles on CO. Preliminary data show that methanol-powered vehicles (both M85 and M100) will emit much more CO than CNG-powered vehicles.

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TABLE 1. *COMPARISON OF EXHAUST* REACTIVITY FOR DIFFERENT FUELS

Fuel Type	Reactivity Factor ^b
Gasoline	1
M85	.56
M85-Low HCHO	.5
LPG (Dual-Fueled)	.85
LPG (Dedicated)	.67
CNG (Dual-Fueled)	.45
CNG (Dedicated)	.36

*Emissions include NMHC, formaldehyde, and methanol, but exclude methane.

^bRelative to gasoline.

Source: Reference 40.

TABLE 2. METHANOL EXHAUST EMISSIONS FROM 1981 AND NEWER AUTOMOBILES BURNING METHANOL

Fuel	Methanol (g/ml) (Range in database)
M85	0.14 ^a - 1.6 ^b
M100	0.33 ^c - 1.7 ^d

^aAverage of 3 lean-burn vehicles (42-CARB 88).

^b50,000 mile projection (2-EPA 87).

^cOne advanced prototype vehicle (1-EPA 89).

^dAverage of emission test results on vehicles fueled with greater than 90 percent methanol (3-EPA 89).

TABLE 3. ESTIMATED NMHC RUNNING LOSS AND REFUELING EMISSIONS
1981+ LIGHT-DUTY VEHICLES - SUMMER

	Running Loss (g/mi)		Refueling Loss (g/mi)
	60 - 86°F	80 - 95°F	
FLEET LDCV (MOBILE4)	0.28	0.59	0.25
M85	0.13 ^a	0.38 ^a	0.1 ^a
M100	0	0	0
CNG - DUAL FUEL 0.14/0.1 ^b	0.23 ^a	0.41 ^a	0.25 ^c
CNG - DEDICATED	0	0	0
LPG - DEDICATED ^d	0 ^e	0 ^e	0 ^e

^aDetermined by multiplying MOBILE4 gasoline projection by ratio of projected M85 vapor/MOBILE4 vapor

^bAssumed to be equal to MOBILE4 emissions with RVP = 8.0 psi

^cAssumes 90% natural gas, 20% gasoline operation.

^dAssumes no fugitive emissions.

^eSmall amount of refueling losses. Data not available.

^fLPG - Dual-fuel will be similar to CNG dual-fuel.

LDCV = Light duty gasoline vehicles

NOTE No emission standard has been established for running losses, originally assumed to be zero

TABLE 4. EVAPORATIVE METHANOL EMISSIONS FROM 1981 AND NEWER
METHANOL-FUELED VEHICLES

Fuel	Methanol (g/mi)
M85	0.02 ^a -0.37 ^b
M100	0.12 ^c

^aTwo advanced prototypes (42-CARB 88).

^b50,000 mile in-use projection (2-EPA 87).

^cOne test (2-EPA 87).

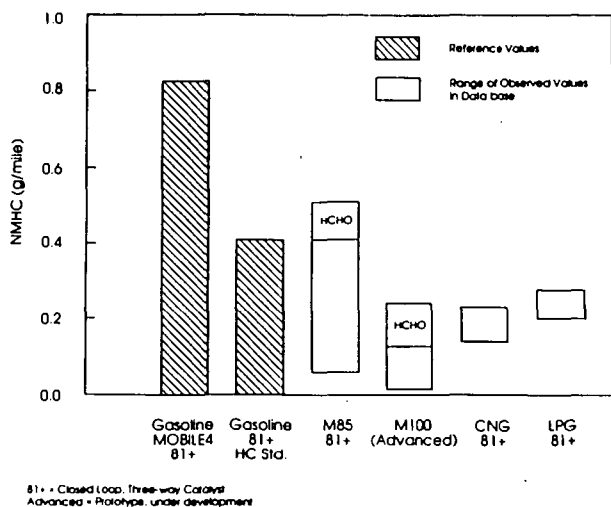


Figure 1. NMHC Exhaust Emissions from Light-duty Vehicles

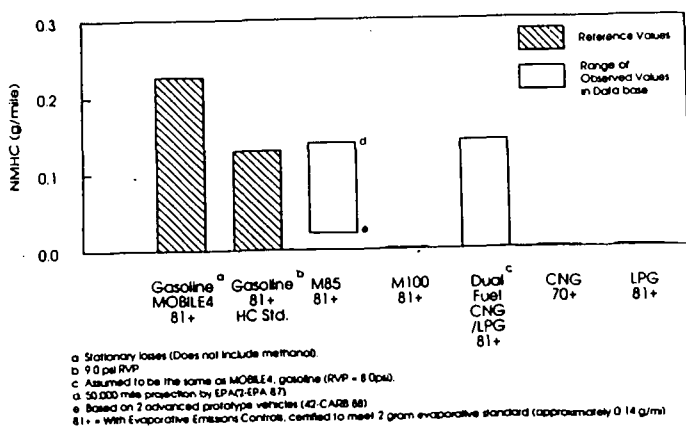


Figure 2. NMHC Evaporative Emissions* from Light-duty Vehicles

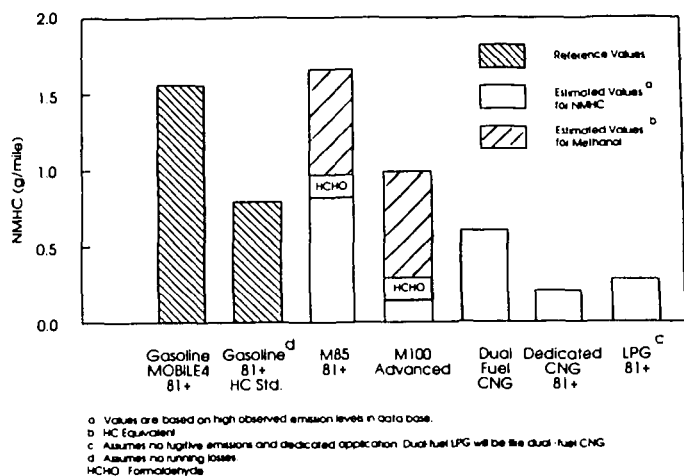


Figure 3. Estimated Total VOC from Light-duty Vehicles (Exhaust plus Evaporative Losses) during Average Summer Weather (60°-84°F)

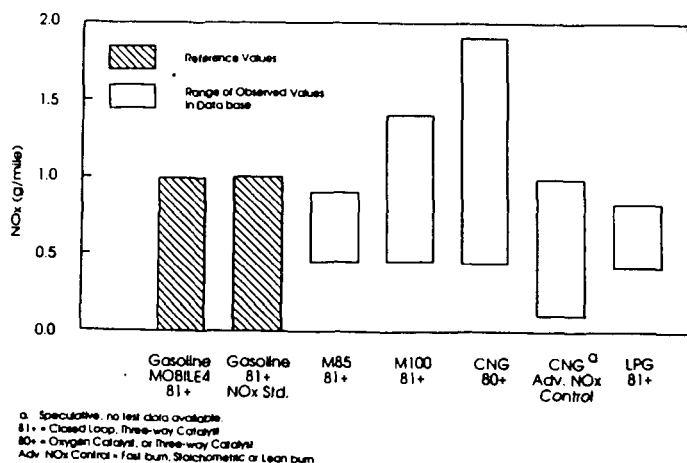


Figure 4. Estimated NO_x Emissions from Light-duty Vehicles

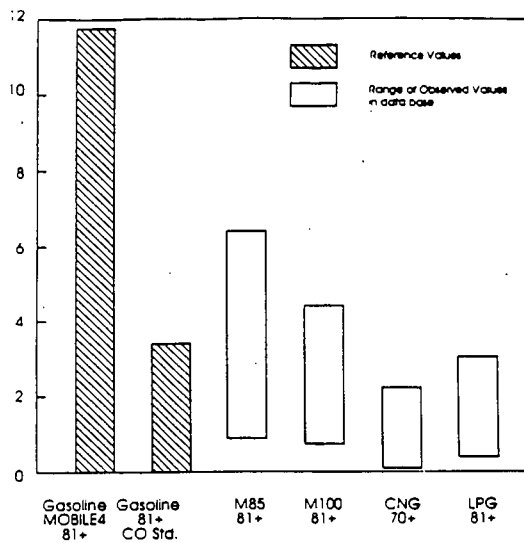


Figure 5. CO Emissions from Light-duty Vehicles

PAH, NITRO-PAH, AND MUTAGENICITY ASSOCIATED WITH DIESEL EMISSIONS MODIFIED BY USE OF A CERAMIC PARTICLE TRAP.

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KEY PHRASES: Heavy-duty Diesel Emissions; Polynuclear Aromatic Hydrocarbon Concentrations; and Mutagenicity Characterization.

INTRODUCTION

Whole diesel exhaust has recently been classified as a probable human carcinogen by the International Agency for Research on Cancer (1) and a potential occupational carcinogen by the National Institute for Occupational Safety and Health (2). Much of the research on the possible health effects of diesel emissions has focused on the particle phase and associated organics, which include many polynuclear aromatic hydrocarbons (PAH) with known biological activity (1,3). The particle-free vapor phase exhaust component has been less extensively studied, with mixed results as to its potential health effects (1). However, this phase has also been shown to contain many of the same types of PAH as found with the particles (1,4,5), some of which could adsorb onto atmospheric particles after leaving the diesel engine.

Various federal regulations have been established concerning allowable levels of oxides of nitrogen (NO_x), hydrocarbons (HC), and particles or total particulate matter (TPM) for on-road heavy-duty diesel emissions for 1991 and 1994 (6). Engine manufacturers have generally been able to meet all of the 1991 standards and the 1994 NO_x and HC standards by making engine modifications. However, it is likely that some type of exhaust aftertreatment device will be required on some engines to meet the 1994 TPM standards, even with the use of low sulfur fuels. One type of device that would likely be used with some types of engines is the uncatalyzed ceramic particle trap. Traps will also be used on buses and on underground mine vehicles. The exhaust must pass through the walls of the trap, which typically results in $\geq 90\%$ removal of particles depending on the type of engine and how it is operated. However, studies with a 1979 heavy-duty diesel engine indicate that HC and vapor phase organics, including PAH, may be only minimally affected by trap usage (4).

This study was designed to assess the effects of an uncatalyzed ceramic particle trap on the regulated and unregulated emissions such as PAH from a late-model, heavy-duty diesel engine. In addition to monitoring effects on NO_x , HC, and TPM, detailed analyses were made of organic material from both the particle and vapor phases for levels of selected biologically active PAH and nitro-PAH as well as mutagenicity. These data were obtained as part of a larger study funded by the Health Effects Institute concerned with obtaining some of the quantitative data necessary for the evaluation of the toxicological impact of employing particle trap technology with modern heavy-duty diesel engines.

EXPERIMENTAL

Engine Operation and Emissions Measurements - A 1988 Cummins Engine Co. LTA10-300 heavy-duty diesel engine was used throughout this study. This turbocharged and aftercooled, direct injection engine met the 1988 federal and California Air Resources Board emissions standards (6) without any exhaust aftertreatment and was considered to be representative of engines on the road in the 1990's. The uncatalyzed ceramic particle trap was a Corning type EX-54 four-piece "quad" (0.305 m long and 0.289 m diameter) with a cell density of 15.5 cells/cm^2 , a wall thickness of 0.43 mm, and a mean pore size of $26 \mu\text{m}$. One batch of AMOCO Premier No. 2 diesel fuel was used, having a cetane index of 48.6, sulfur content of 0.316 wt %, and aromatics content of 22.0 vol %. The engine oil used was Cummins Premium Blue (SAE 15W-40).

Emissions with and without the trap were collected with the engine operating at two rated speed (1900 rpm) modes of the EPA 13-mode steady-state cycle, i.e., modes 9 (75% load)

and 11 (25% load). These modes were selected as being representative of on-highway operation, having similar emission characteristics to data for transient operation of this engine, and having exhaust temperatures below those at which trap self-regeneration should occur. (However, later testing indicated that some trap self-regeneration was occurring at mode 9 due to higher trap temperatures.)

Levels of HC, NO_x, and nitric oxide (NO) were determined in the raw exhaust using a Beckman flame ionization detector HC analyzer and a chemiluminescence NO/NO_x analyzer, respectively. A portion of the raw exhaust with or without (baseline) trap usage was split-off and injected into a dilution tunnel. The volume dilution ratio was calculated by the ratio of measured raw NO_x to measured dilute NO_x.

Portions of the dilution tunnel flow were diverted simultaneously into one of two sampling systems, i.e., a 47-mm filter system used for determination of TPM and component levels [soluble organic fraction (SOF), sulfate, and solids (SOL)] and a 508 x 508-mm filter system used for collection of particle and vapor phase materials for chemical and biological characterization. Both systems contained the same type of Teflon-coated woven glass fiber filters (Pallflex TX40H120-WW). A container with 40 g XAD-2 resin was located immediately behind the 508 x 508-mm filter to collect semi-volatile organics (XOC).

In order to minimize dilution tunnel sampling variability (7), the filter face temperature under all engine operating conditions was maintained at 45 ± 2°C by allowing the dilution ratio to vary as ambient temperatures (tunnel inlet temperatures) varied for different runs. In order to also minimize the potential for artifact formation, sampling times were held at ≤60 minutes and filter face nitrogen dioxide levels at <5 ppm.

Levels of TPM on the 47-mm filters were determined gravimetrically. Soluble organic fraction levels were determined gravimetrically after filter extraction for four hours with dichloromethane in a Soxhlet apparatus. Sulfate levels were then determined using ion chromatography. Any remaining mass on the filters was considered to be the SOL component. Levels of SOF and XOC associated with the 508 x 508-mm filters and XAD-2 resin, respectively, were determined after Soxhlet extraction for 24 hours with dichloromethane. The mass of each extract was determined gravimetrically by allowing the SOF or XOC to dry on a known mass of Florisil.

Compound Quantitation - The PAH and nitro-PAH selected for quantitation by high performance liquid chromatography (HPLC) were: benz[a]anthracene; benzo[b]fluoranthene; benzo[k]fluoranthene; benzo[a]pyrene; chrysene; fluoranthene; 7-nitrobenz[a]anthracene; 6-nitrochrysene; 3-nitrofluoranthene; 2-nitrofluorene; 1-nitropyrene; and pyrene. These compounds were selected for quantitation due to their known or suspected health effects (1,3), their known occurrence in diesel exhaust (1,3-5,7), the availability of suitable reference materials, and the ability to resolve them from apparent interferences in our quantitation procedure (7).

Separation of the SOF or XOC samples and compound quantitations were conducted using the scheme presented in Figure 1. Quantitation was accomplished by using internal standard method calculations using 2-chloroanthracene and comparison to the National Institute for Standards and Technology standard reference material 1647a as the reference (calibration) standard for PAH and standard reference material 1597 for the nitro-PAH calibration standard. Minimum detection levels (MDL) were determined using dilutions of the calibration standards, with MDL considered to be those quantities injected on the HPLC having a minimum of three times the background noise and areas which could be reproducibly measured. As 1-nitropyrene and 1,3-dinitropyrene coeluted by the method noted for nitro-PAH quantitation, the results are presented as "1-nitropyrene + 1,3-dinitropyrene" levels.

Mutagenicity Analyses - Mutagenicity associated with the SOF and XOC samples was determined using a small dish modification of the microsusension version of the *Salmonella typhimurium*/microsome mutagenicity bioassay (8-10). Samples for the mutagenicity bioassays were prepared by dissolving a known mass of SOF or XOC in a known volume of dimethylsulfoxide. All samples were tested with tester strain TA98 ± S9 metabolic activation and retested with the nitroreductase-deficient tester strains TA98NR and TA98-1,8-DNP₆ if

responses were detected with TA98-S9. The appropriate solvent, positive control, genotype, sterility, and viability controls were conducted with each test (8).

Data Analysis - As data from two to six test dates per mode and condition were available for analysis, this study was considered to have an unbalanced design with test dates as replicates (11). There was an unequal number of samples per replicate date for each mode and condition; therefore, a mean of means was calculated for most emission parameters, except as noted below. A two-way analysis of variance (ANOVA) was used to compare mode (9 and 11) versus condition (baseline and trap) means at a significance level of 0.05. Where a significant interaction term occurred, a one-way ANOVA was used.

Exceptions had to be made to the statistical analysis procedures (as described above) for many of the PAH and nitro-PAH measurements and the mutagenic activity data. The latter could not be analyzed statistically because the revertants/ μg value was calculated using a power function model that was dependent upon the interaction of both the slope and the y-intercept (12). The frequent occurrence of "less than MDL" values in the compound quantitation data sets meant that mean daily values typically could not be calculated. Therefore, mean of median values were determined for each mode/condition for use in the ANOVA. If the majority of values for any test date was above the MDL, then that median value was used for that test date. If 50% or less of the values for each test date were above the MDL, then a value was assigned to each "less than MDL" value for median determinations using several different techniques. The technique that appeared to have the least scattering of residuals was the replacement of the "less than MDL" value with "MDL/2". All ANOVA results reported in this paper involving mean of median analyses were conducted using this technique.

RESULTS AND DISCUSSION

This paper summarizes the results of one of the few large heavy-duty diesel emission sample sets comprising multiple test dates and engine operating modes and conditions. It also represents the first detailed quantitative study specifically of biologically active PAH and nitro-PAH compounds associated with both the particle and vapor phase exhaust from a late-model diesel engine.

Gaseous and Particle-Associated Emissions - Baseline emissions from this 1988 Cummins heavy-duty diesel engine were notably different from older engines tested at Michigan Technological University (MTU) (4, 7, 12). Not only were TPM levels much lower with the 1988 engine, but the proportion of the HC recovered on XAD-2 resin (as XOC) compared to the filters (SOF) was greatly increased.

As indicated in Table 1, use of the trap with the Cummins engine had no significant effects on NO_x (or NO) concentrations at either mode but exerted its effects on the HC (and XOC), TPM, and TPM-associated exhaust components. The greatest effects were found with reductions in SOL concentrations of up to 95%, as this type of control device acts primarily to physically remove this material via engine mode-independent mechanisms. These results are in agreement with previous studies with other types of diesel engines (4, 12-14). The reductions in SOF can be related both to some significant effects on HC levels (at mode 9) and to the decrease in SOL surface sites for adsorption of organic material found at both modes. Sulfates were also decreased with trap use, although a significant change was found only at mode 9. These combined reductions resulted in 92% and 80% TPM-removal efficiencies at modes 9 and 11, respectively. There were consistent mode-dependent differences in trap effects on HC and XOC concentrations, with the latter representing a portion of the vapor phase HC. The significant changes in HC and XOC concentrations (reductions of 43 and 67%, respectively) at mode 9 but virtually no change at mode 11 were most likely related to differences in exhaust temperature between the two modes. The higher trap temperature at mode 9 (450°C) versus mode 11 (325°C) probably resulted in greater oxidation of HC within the trap and, thus, lower HC and XOC levels after the trap. A high proportion of the recoverable HC were still vapor-phase associated (as XOC) even with trap use at mode 9.

Mutagenicity Effects - A summary of the trap and mode effects on TPM and XOC-associated mutagenic activities is presented in Table 2. All detected mutagenicity was direct-acting in nature, regardless of mode, trap usage or particle or vapor phase association, as has

typically been found for diesel exhaust-associated organics. However, in contrast to previous studies with older model engines at MTU (4,12) and elsewhere (13,14) with this same type of control device, trap use with this 1988 engine did not produce increases in TPM-associated mutagenic activities (revertants/ μg). The 93% and 86% decreases in exhaust mutagenic concentrations (revertants/ m^3) at modes 9 and 11, respectively, were similar to the reductions found in TPM and SOF concentrations (Table 1). Similar to our earlier study with a 1979-model heavy-duty diesel engine (4), about 50% of the recovered mutagenicity (TPM plus XOC) was associated with the XOC. Unlike the earlier study, no mutagenicity was detected at either mode with trap usage with this 1988 engine. Differences in engine configurations and operation, emissions levels (such as for NO_x and NO), and sampling procedures may account for these differences in trends between the present and previous studies.

The responses of the two nitroreductase-deficient tester strains (Table 2) indicated more of the observed TPM-associated activity was due to nitro-substituted compounds under baseline than under trap conditions. The same general relationship was found between baseline TPM and XOC-associated mutagenicity, with the exception of the TA98-1,8-DNP₆ responses at mode 11. This likely indicates differences in the proportion of nitro-substituted compounds in the particle versus vapor phases under baseline conditions and, perhaps, lower levels of these mutagenic compounds with the trap.

PAH and Nitro-PAH Constituents - The ranges of PAH and nitro-PAH concentrations found for each operating condition with and without the trap are presented in Table 3. The baseline particle-associated concentrations for many of these compounds are lower than those in our earlier study with an older engine using similar quantitation techniques (7), in part due to the lower TPM levels with the 1988 engine. However, comparisons to literature values for other studies are difficult due to differences in engines, operating and sampling conditions, and analytical procedures. As noted in the "Experimental" - Data Analysis" section, presentation and analysis of most of these data sets were complicated by the presence of "less than MDL" values. With the exception of the fluoranthene TPM and XOC, pyrene TPM and XOC, and chrysene TPM data sets, all of the PAH and nitro-PAH data were analyzed using procedures to estimate medians. Even so, several of these ANOVA comparisons did result in significant differences between either trap use or mode.

Trap usage had significant effects on fluoranthene, benzo[b]fluoranthene, and 7-nitrobenzo[a]anthracene TPM-associated concentrations at mode 9 and pyrene and 7-nitrobenzo[a]anthracene at mode 11. With the exception of pyrene, the concentrations of these compounds decreased with trap use. The general trend, which was more pronounced at mode 9 than 11, was for decreases in concentrations with use of the trap. Pyrene was the only compound with significant differences in concentrations between modes, although the general trend was for higher concentrations at mode 11 than at mode 9. This trend is in agreement with the higher exhaust temperatures and greater reductions in HC and SOF found at mode 9.

In contrast, the trap was found to have no significant effects on vapor phase compound concentrations. There were significant differences in fluoranthene, 6-nitrochrysene, and 1-nitropyrene + 1,3-dinitropyrene concentrations between modes, with the lower concentrations again found at mode 9.

Several PAH, such as the benzo[fluoranthenes and benzo[a]pyrene, were found almost exclusively with particles and some such as fluoranthene and pyrene were proportionally much higher in the vapor phase; these associations were generally in accordance with the known vapor pressures for these compounds (5,15). However, all of the other PAH and nitro-PAH were detected in the vapor phase emissions of this engine with or without the particle trap; a general trend was for an increase in the vapor phase concentration of many of these compounds with trap usage.

CONCLUSIONS

This uncatalyzed ceramic particle trap had consistently significant effects (up to 95% reductions) on TPM and component concentrations regardless of engine operating condition. The PAH and nitro-PAH particle-associated concentrations generally were reduced with trap use, but this was not necessarily true with the vapor phase-associated concentrations. Some

compounds appeared to shift from the particle to vapor phase with the trap. Mutagenicity associated with the particles was generally unaffected with the traps, but mutagenic concentrations decreased due to decreases in particle concentrations. All detectable vapor phase mutagenicity was eliminated with trap use. Therefore, this type of emissions control device appeared to reduce particle concentrations from this late-model heavy-duty diesel engine without increasing particle-associated levels of biologically active PAH or nitro-PAH or mutagenicity. However, the impact of increases in certain types of compounds in the vapor phase should be investigated further.

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Table 1. Effect of a Ceramic Particle Trap on Emissions.

Parameter ^a	Mode 9		Mode 11	
	Baseline (mg/m ³)	Trap Effect (% change)	Baseline (mg/m ³)	Trap Effect (% change)
NO _x	1,390	+1.4	430	+8.8
HC	41.8	-43*	87.2	-6.0
TPM	27.5	-92*	34.0	-80*
SOF	4.40	-89*	7.95	-75*
SOL	17.8	-95*	23.3	-86*
SO ₄	5.08	-80*	2.77	-40
XOC	28.0	-67*	42.8	-4.0

^aNO_x = oxides of nitrogen; HC = hydrocarbons; TPM = total particulate matter; TPM components - SOF = soluble organic fraction, SOL = solids, and SO₄ = sulfates; and XOC = XAD-2 resin organic component.

*Mean significantly different from comparable baseline mean (p ≥ 0.05).

Table 2. Effect of a Ceramic Particle Trap on Mutagenic Activity.

Mode/ Condition	Component ^a	Mutagenic Activity, TA98-S9 ^b		% of TA98-S9 Response		
		Revertants/μg	kRevertants/m ³	TA98+S9	TA98NR-S9	TA98DNP-S9 ^c
9 Baseline	TPM	0.60	20.6	55	15	15
	XOC	0.67	18.6	105	55	64
9 Trap	TPM	0.42	1.39	92	60	46
	XOC	NR ^d	NR	NR	NT	NT
11 Baseline	TPM	0.68	27.3	72	24	24
	XOC	0.56	24.7	75	40	13
11 Trap	TPM	0.39	3.96	78	60	61
	XOC	NR	NR	NR	NT	NT

^aTPM = total particulate matter; XOC = XAD-2 resin organic component.

^bRevertants/μg based on power function model analysis of dose-response data.

^cTA98DNP = TA98-1,8-DNP₈.

^dNR = no response above spontaneous revertant levels; NT = not tested as no activity with TA98-S9.

Table 3. Range of PAH and Nitro-PAH Concentrations.

Compound	Concentration Range (ng/m ³) ^a							
	9 Baseline		9 Trap		11 Baseline		11 Trap	
	TPM	XOC	TPM	XOC	TPM	XOC	TPM	XOC
Benz[a]anthracene	24-310	82-190	<1.1-76	<0.37-48	120-310	160-280	88-220	8.1-400
Benzo[b]fluoranthene	6.1-190	≤85 ^e	<0.97-52 [*]	<0.16-11	16-68	<4.6 ^d	<1.2-95	<0.96-180
Benzo[k]fluoranthene	<1.7-24	<2.8 ^d	<2.4 ^d	<3.0 ^d	<1.9-6.7	<4.6 ^d	<1.9 ^d	≤34 ^e
Benzo[a]pyrene	<1.4-22	<2.0 ^d	<0.14-1.3	<3.1 ^d	<1.3-5.2	<4.8 ^d	<1.4-13	<2.9 ^d
Chrysene ^b	24-340	1.4-49	4.6-110	<0.35-45	23-360	<0.31-40	12-230	<8.7-450
Fluoranthene ^c	520-23,000	1,300-2,200	57-2,100 [*]	88-3,000	410-780	2,100-3,500	190-2,900	730-6,500
Pyrene ^c	8.1-84	120-590	13-380	99-580	69-130	390-650	260-790 [*]	690-1,600
7-Nitrobenz[a]-anthracene	0.60-3.9	2.5-9.5	0.14-2.1 [*]	≤0.34-60	<4.0-7.2	14-63	≤1.3-18 [*]	<1.1-14
6-Nitrochrysene	2.1-13	1.7-2.5	<0.88-4.3 ^e	0.69-8.0	≤0.27-0.76	≤0.65-12	1.6-11	<0.95-7.0
3-Nitrofluoranthene	≤1.4 ^e	<2.8 ^d	<2.1 ^d	0.016-13	0.17-2.0	3.4-12	≤0.66-4.2	<1.1-7.5
2-Nitrofluorene	5.3-23	2.1-9.9	<2.0-21	<0.12-69	5.2-5.6	1.8-28	7.4-18	≤2.1-26
1-Nitropyrene + 1,3-Dinitropyrene	<2.1-21	<2.7 ^d	0.86-25	0.13-45	<3.8-38	12-190	<3.8-47	<1.1-65

^aTPM = total particulate matter; XOC - XAD-2 resin organic component.^bValues above minimum detection limits (MDL) for TPM samples.^cValues above MDL for all TPM and XOC samples.^dNo values > MDL.^eAbove MDL value for one sample only.^{*}Mean significantly different from comparable baseline mean (p = 0.05).

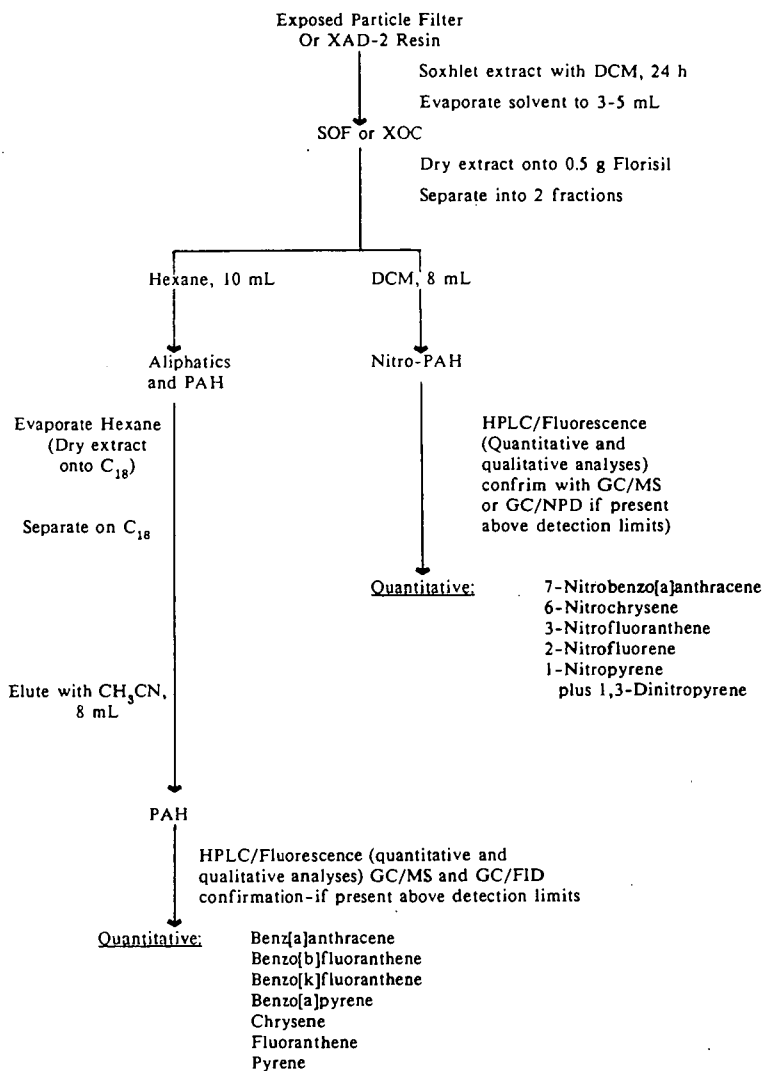


Figure 1. Flow chart for the extraction of SOF and XOC and compound quantitation. DCM = dichloromethane; SOF = soluble organic fraction; XOC = XAD-2 resin organic component; PAH = polynuclear aromatic hydrocarbons; CH₃CN = acetonitrile.

IMPACT OF FUEL CHOICE ON COMPARATIVE CANCER RISK OF EMISSIONS

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ABSTRACT

Incomplete combustion of fuels as an energy source results in the emission of products which are mutagenic in short-term genetic bioassays and carcinogenic in animals. Humans occupationally exposed to the incomplete combustion products from certain fuels have an elevated relative risk of cancer. Until recently, it has not been possible to compare the relative cancer risk of emissions from the combustion of various fuels. The combustion emissions from a wide variety of fossil fuels, synthetic fuels, vegetative fuels, synthetic chemicals, and mixed wastes have been characterized for with respect to their comparative genotoxic and tumor initiating activity. This paper evaluates the comparative cancer risk of various fuels in our current data base and identifies critical data gaps in our understanding of the comparative cancer risks from using alternative fuels.

INTRODUCTION

Fossil fuel related mixtures were the first chemicals recognized as human and then animal carcinogens (1). Coal combustion products, chimney soots, and coal tars were the first fossil fuel derived chemical carcinogens studied. Evaluation of the weight of evidence for the carcinogenic risk to humans of many fuel related mixtures has been conducted by the International Agency for Research on Cancer (IARC)(2). Based on these evaluations soots, coal-tars, and shale oils are classified as carcinogenic to humans (Group 1). Carcinogenicity in animals, with limited evidence in humans, has led to diesel engine exhaust and occupational exposures in petroleum refining to be classified as probably carcinogenic to humans (Group 2A). For several other petroleum based fuels (e.g., unleaded gasoline, marine diesel fuel, and residual fuel oils) and their combustion products (e.g., gasoline engine exhaust) the IARC has classified these mixtures as possibly carcinogenic to humans (Group 2B) based principally on the evidence for carcinogenicity in animal studies. Clearly, human exposure to a number of fossil fuel products, especially the combustion products, presents a potential cancer risk.

Vegetative carbon, from non-fossilized plants, was probably the first carbon source used as a fuel. Unfortunately, with the exception of tobacco combustion products, few studies have been conducted to evaluate the human cancer risk from vegetative carbon sources. A lung cancer mortality study in China in a non-smoking female population exposed to high concentrations of indoor burning of either "smoky" coal, "smokeless coal" or wood show that the lung

cancer mortality rates are highest in the populations using the smoky coal (3).

Alternative coal and shale derived fuels and products associated with technologies such as coal liquefaction were evaluated for genotoxicity, carcinogenicity, and other toxicological effects by industry and the U.S. Dept. of Energy (4). In many cases these alternative "synfuels" were compared to conventional petroleum-based fuels. No studies have been conducted on the cancer risk from alternative fuels such as synthetic chemical fuels, oxygenated petroleum fuels and alcohol based fuels.

HUMAN EXPOSURE

The most significant human exposures from fuel use are the air pollution exposures resulting from fuel related combustion emissions from transportation, heating and other area sources (5). Combustion emissions are a complex mixture of gases, condensable organics, and particles. The particles vary from classical submicron carbonaceous soot particles with condensed organics, and inorganic particulate matter to a mixture of condensable organic matter with almost no carbonaceous soot. The condensed or adsorbed organic, often referred to as "tar" in the earlier literature contains the polycyclic organic matter (POM) which induces tumors in animals, mutations in cells, and has been clearly implicated in epidemiological studies as a human carcinogen (5,6).

Polycyclic organic matter (POM) is a general term referring to a complex organic mixture of polycyclic aromatic compounds including many diverse classes of hydrocarbons (e.g. polycyclic aromatic hydrocarbons, PAH), substituted aromatic hydrocarbons (e.g., nitrated-PAH), heterocyclic aromatic compounds (e.g., aza-arenes). Incomplete combustion products, however, also contain gaseous chemicals which are carcinogenic such as benzene, aldehydes, and alkenes (e.g., 1,3-butadiene) and semi-volatile organic compounds which have not been well characterized either chemically or toxicologically (6).

The complexity of the POM emissions, estimated to contain thousands of chemicals, has precluded the quantitative cancer risk assessment of these emissions based on analysis of the components. Since human exposure to these POM emissions occurs as the whole complex mixture, both qualitative, weight of evidence, assessments (2) and quantitative assessments (7) of the human cancer risks have been based on either the whole emissions or the POM component.

CANCER POTENCY AND RISK

Three different approaches have been taken to the quantitative assessment of human cancer risk from fuel related mixtures: 1. Low dose extrapolation of human cancer risks at relatively higher occupational exposures (8), 2. Extrapolation of chronic animal cancer studies to human risk using cross species extrapolation methods (7) and 3. Extrapolation from relative tumor potency data in animals to relative cancer risk in humans based on the comparative

potency method. The first method, relying only on human data, has been the most extensively used for fuel related mixtures (e.g., coke oven emissions (8)), however there is not adequate human data to assess the potential cancer impact of either the currently used conventional fuels (e.g., petroleum derived) or the possible alternative fuels that may be used in the future. The second method, relying on chronic animal cancer data, is also severely limited by the available chronic animal data. The third method, relies on a comparative potency data base for a series of combustion related POM mixtures which have been compared in animal tumor assays and short-term bioassays to the relative human lung cancer risk in a series of related POM mixtures.

The comparative potency method for cancer risk assessment of these complex POM emissions is based on the constant relative potency hypothesis. This method was developed and tested using human lung cancer unit risk estimates, animal tumorigenicity data, and short-term mutagenesis bioassay data (9,10). These mixtures included the extractable organic emissions from coke ovens, roofing coal tar pots, cigarette smoke and automotive emissions. The comparative potency method is based on the hypothesis that there is a constant relative potency between two different carcinogens across human and bioassay data. The mathematical expression for the constant relative potency model is the following:

$$\frac{\text{bioassay potency of } \frac{\text{carcinogen}_a}{\text{carcinogen}_b}}{\text{human cancer potency of } \frac{\text{carcinogen}_a}{\text{carcinogen}_b}} = (k) \text{ constant}$$

The constant relative potency assumption is implicit in any comparison which utilizes the relative toxicity of two substances in animals to estimate their relative toxicity in humans. This constant relative potency assumption is an experimentally testable hypothesis, if the relative potency of two mixtures or components in one bioassay (e.g., humans) can be determined and compared to the relative potency in a second bioassay. The test of this hypothesis is whether there is a constant relationship (k) between the relative potencies in the two bioassay being compared. This hypothesis was tested for three complex organic emissions from a coke oven, roofing coal tar pot and cigarettes by using the human lung cancer data from epidemiological studies of humans exposed to these emissions and comparing the lung cancer unit risk to the potency of these emission sources in a series of bioassays (7,8).

The first application of this method to estimation of the human lung cancer unit risk was for the POM associated with diesel particle emissions (9). In this study, the comparative human data used was for POM from coke ovens, roofing coal tar emissions and cigarette smoke. Evaluation of a battery of bioassay demonstrated that across these three human carcinogens, the constant relative

potency hypothesis could be validated for the mouse skin tumor initiation potency. This is shown in Table 1 by the correlation between the human lung cancer unit risk and the tumor initiation potency for the three human carcinogens.

Table 1.
CANCER UNIT RISK ESTIMATES FOR THE HUMAN COMPARATIVE EMISSIONS

EMISSION SOURCE	HUMAN CANCER POTENCY ^a lifetime risk/ug EOM/m3	TUMOR POTENCY ^b pap/mouse/mg EOM
Coke Oven	9.3×10^{-4} (1.0) ^c	2.1 (1.0)
Roofing Tar (Coal)	3.6×10^{-4} (0.39)	0.41 (0.2)
Cigarette Smoke	2.2×10^{-6} (0.0024)	0.0024 (0.0011)

^aLifetime cancer unit risk per microgram extractable organic matter (EOM) per cubic meter determined from human epidemiological data(9)

^bPapillomas per mouse per mg of EOM ^cRelative potency shown in ().

Using this methodology the lung cancer unit risk has been estimated for the following POM emission sources as shown below:

Table 2:
CANCER UNIT RISK ESTIMATES DETERMINED BY THE COMPARATIVE POTENCY METHOD^a

EMISSION SOURCE	POM UNIT RISK (lifetime risk/ug organic matter/m3)	PARTICLE UNIT RISK (lifetime risk/ug particulate matter/m3)
Automobile-Gasoline Catalyst Non-catalyst	12.0×10^{-5}	5.1×10^{-5} 1.6×10^{-5}
Automobile-Diesel ^b Trucks-Diesel	23.0×10^{-5} 0.7×10^{-5}	2.6×10^{-5} 0.2×10^{-5}
Woodstoves	2.9×10^{-5}	1.0×10^{-5}
Heating Fuel Oil		0.9×10^{-5}

^aMouse skin tumor initiation bioassay potency was used as the comparative bioassay.

^bAverage of the unit risk estimate for three light-duty diesel vehicles(9).

The validity of this constant relative potency hypothesis may

depend on the chemical nature of the mixtures being compared as well as the similarity of those mixtures. Comparison of the mutagenic potency of a series of POM from diesel and gasoline vehicle emissions in Salmonella typhimurium with the tumorigenic potency showed high correlations both between the two bioassay and with the concentration of nitrated PAH and PAH in the POM mixture (11).

IMPACT ANALYSIS OF CANCER RISK

Human cancer risk from exposure to a fuel or its combustion products is dependent on the extent of exposure (e.g., dose or exposure concentration x time of exposure) and the potency of the carcinogen (e.g., cancer risk/unit dose) among other factors (e.g., individual susceptibility). It is important to develop tools to predict the potential impact of cancer risk in the absence of human exposure assessment data so that such assessments can be conducted prior to the widespread introduction of new fuels or technologies into commerce. By combining source emission studies with mutagenicity and tumorigenicity bioassay studies of the combustion source emissions, we are able to estimate the relative impact of various alternative fuels as shown in Table 3.

Table 3.
COMPARATIVE MUTAGENIC AND TUMORIGENIC EMISSION FACTORS

SOURCE	MUTAGENIC EMISSION FACTOR (rev/kg fuel x 10 ⁵) ^a	TUMORIGENIC EMISSION FACTOR (pap/mouse/kg fuel) ^b
RESIDENTIAL HEATING wood oil	50 1	83 3
AUTOMOTIVE SOURCES diesel vehicles gasoline non-catalyst gasoline catalyst	40 10 1	240 20 2

^aRevertants (rev) measured using the Salmonella typhimurium mutagenicity bioassay

^bPapillomas (pap) measured using the mouse skin tumor bioassay

This analysis of the potential impact of sources based on the relative bioassay potency expressed per kg fuel consumption shows even more clearly the comparative differences between these fuels and sources. In this analysis the potential human exposure is assumed to be directly related to the emission rate. Although wood combustion emissions are less potent as carcinogens than many of fossil fuel combustion emissions, the high organic emission rate of woodstoves results in a significant impact when both potential exposure and potency are combined.

To examine the impact of fuel within one combustion source, we have conducted fuels comparison studies by operating motor vehicles, with different qualities and sources of diesel fuels. In several independent studies of diesel fuels we consistently found that alterations in the combustion source (engine type or operating conditions) resulted in greater differences in the mutagenic emission rates than differences in fuel quality or source (e.g. shale derived vs petroleum) (12).

Unfortunately few such fuel comparison studies have been conducted with gasoline vehicles, residential heating sources, waste burning and other significant fuel combustion sources. New oxygenated fuels being widely introduced into vehicle fleets across urban areas are expected to result in reduced POM emissions and reduced risk. However, no studies of the potential genotoxicity or tumorigenicity of these emissions have been conducted. The combustion of synthetic chemicals such as plastics mixed in municipal and hospital wastes and hazardous waste is the least well characterized with respect to the biological activity or potential cancer impact of the complex mixtures emitted from these sources.

DISCLAIMER

The research described in this paper has been reviewed by the US EPA and approved for scientific publication. The contents do not necessarily reflect the views and policies of the Agency.

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THE CLEAN COAL INITIATIVE: AN APPROPRIATE
RESPONSE TO COMPLEX ENVIRONMENTAL ISSUES

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INTRODUCTION

Little effort is required to confirm the importance of coal as a fuel source for meeting the increasing demands for energy both in the U.S. and internationally. Performing such an exercise would establish that the U.S. should be preparing to use twice as much coal representing an increase in its share of total U.S. energy consumption from today's 34 percent to about 40 percent over the period between now and the middle of the next century.

The challenge is not whether coal will be used, but to assure its use is accomplished in the most efficient way and with the least impact on the environment. Environmental requirements have joined cost reduction as the primary driving forces pushing coal technology development in new directions. A responsive and successful technology development program is essential to realizing sustainable economic and environmental development of our coal reserves on a domestic as well as on a global basis.

TECHNOLOGY DEVELOPMENT

It is an incontrovertible fact that the uncontrolled burning of coal will release into the environment carbon dioxide (CO_2), sulfur dioxide (SO_2), oxides of nitrogen (NO_x), particulate matter, and ash. The relative amounts of these emissions will be determined by the amount of coal burned and the effectiveness as well as the degree to which some type of controls are used. The CCTs create the capability to utilize coal as an energy resource with attendant minimal emissions of these pollutants.

Suitably developed CCTs thus provide a unique opportunity to concurrently satisfy two national goals. These are to:

- (a) Continue the substantial progress made in emission reductions by resolving the conflicts between coal use and environmental protection.
- (b) Meet the rapidly approaching electricity capacity challenge with reduced cost and increased productivity.

Recognizing that in the U.S. 85 percent of all coal produced provides about 60 percent of the Nation's electricity, development of CCT is pursuing two primary objectives. These objectives are to:

- (a) Provide an array of retrofittable emission control options which satisfy, at reduced cost the void between present physical coal cleaning capabilities and flue gas scrubbing for existing powerplants.
- (b) Provide improved coal utilization alternatives for both repowering and new plant application. Of particular significance are fluidized-bed combustion and coal gasification/combined cycles which combine high levels of emission and effluent control with reduced cost and improved efficiency.

On a global basis, the importance of this linkage between technology development, electricity, coal utilization, and the environment was underscored by the Conservation Commission of the World Energy Conference which stated that:

- (a) Future global economic growth particularly in the developing regions will be associated with very large increases in electricity growth.
- (b) Coal which provides over 80 percent of the world's nonrenewable energy resource will become increasingly prominent in the coming decades. Worldwide coal usage is expected to exceed the use of petroleum during the first decade of the next century and to continue its preeminence during at least the following 50 years.
- (c) Technological innovation to support this expansion in coal use will be essential for the world's economy and its environment.*
- (d) The geopolitical importance of coal and the technology for its use are also likely to increase substantially since the bulk of the world's coal resources is controlled by the U.S., U.S.S.R., and China.

*Underlining emphasis added.

These assessments reflect the unique opportunity that low cost, widely distributed coal has to become the source of a versatile, highly available energy form--electricity. Electrification has become a global economic imperative. Growth in electric power production is a major consideration in global energy and environmental projections.

While in the past, the pace of the required advancements in coal utilization technology has proceeded at a acceptable rate, new constraints including escalating capital costs, declining construction productivity, new licensing requirements and growing environmental concerns has shown that a fundamental change in the development of coal-fired technology is required. A principle now being used to guide CCT development is that sustained environmental improvement can only be realized when both emission and cost reduction are achieved concurrently, not pitted against each other.

CLEAN COAL TECHNOLOGY DEVELOPMENT

The importance of these factors in developing an appropriate strategy for the use of the immense coal reserves of the U.S. has been recognized. Positive steps have been taken to ensure that new concepts for the utilization of coal are available to respond to future needs.

The Clean Coal Initiative is one of the largest technology development efforts now underway in the Department of Energy (DOE). The level of funding responds directly to the strategic importance of coal in the U.S. economy and

the international marketplace and recognizes the need to develop solutions for the problems (economic and environmental) associated with meeting increased demand for this source of energy.

It is an accepted fact that the present and near term future of coal as an energy source depends upon continued advances in coal utilization technology.

The CCT Demonstration Program itself is a technology development effort jointly funded by the Government and industry. In this program, the most promising of the advanced coal based utilization technologies are being moved into the marketplace through demonstration. The demonstration effort is at a scale large enough to generate all data needed by the public sector to judge the commercial potential of the processes being developed. As a goal, the program will make available to the U.S. energy marketplace a number of advanced, more efficient, and environmentally responsive coal utilization technologies. These technologies will reduce or eliminate the economic and environmental impediments that limit the full use of coal.

The program as planned, currently consists of five phases (rounds). It is entering the fourth of the five planned phases. The first three phases were competitive solicitations in which 35 projects with an estimated total cost of \$3.7 billion were selected. Of these projects, 29 have been started under the terms of cooperative agreements between the participants and the Government and negotiations are in progress on the remaining six. Of particular importance to DOE in these projects is the level of financial participation of the private sector. Although the U.S. Congress, in its guidance to the program, requires that such participation be a minimum of 50 percent, the participants are providing 63 percent of the funds for the projects in the cooperative agreements signed to date.

The fourth phase of the program has been initiated. The fourth competitive solicitation was released on January 17, 1991, with additional projects to be selected in 1991. Funding in the amount of \$600 million each for phases IV and V have been appropriated. When the five phases of the program are completed, two major objectives will have been accomplished:

1. A wide range of more efficient, environmentally responsive coal utilization processes will be available for use that will accommodate almost every combination of specific site requirements.
2. Acid rain precursor emissions will be significantly reduced and significant reductions will have been made in the amounts of CO₂ generated as a result of using coal as an energy source.

WHAT ARE CLEAN COAL TECHNOLOGIES?

This description of something called the CCT Program has little meaning in the absence of some idea about what CCTs are and what type of benefits can be achieved through their use.

The term "clean coal" in itself is to some a conflict in terms. The image of coal certainly is not consistent with "clean." When CCTs are discussed, these

discussions are about advanced coal based utilization systems that offer significant benefits when used for power generation, pollution control, or the conversion of coal into other alternate energy products.

First in the area of power generation, the characteristics of these technologies such as improved thermal efficiencies, modular construction, improved environmental performance, fuel flexibility, repowering capability, etc., will help the power industry accommodate a time of significant change caused by such factors as regulatory reform, uncertain growth in power demand, environmental pressures, and now competition from Independent Power Producers and Cogenerators.

Second, the name clean coal emphasizes its role in pollution control. In this case, most generally through a retrofit application, the technologies can directly remove SO_2 and NO_x acid rain precursors and substantially reduce the amount of CO_2 generated when combusting coal. The specific type and amounts of pollutants removed will be a characteristic of the individual process. It should be noted also, however, that some CCTs (e.g., Pressurized Fluidized-Bed Combustion (PFBC), Integrated Gasification Combined Cycle (IGCC) have the ability to remove these pollutants while at the same time increasing the power output of the facility itself from 50-150 percent.

In the third case, CCTs can create the opportunity to produce coal derived liquid fuels to replace oil and gas in some applications. This capability will permit coal to have a greater role in providing energy to the industrial, commercial, and transportation sectors.

IMPACT OF THE PROGRAM

With the CCT Demonstration Program implemented to only 60 percent of its proposed full potential, it is premature to evaluate the degree to which it will achieve its objective of making available the technology that will permit the increased use of coal in an environmentally acceptable manner. However, it is not premature to examine the program's accomplishments to date to see how well it is progressing toward that objective.

Thirty-five projects representing ten major categories of technology are in various stages of design, construction, and operation. These projects represent a cost shared effort of private industry and government to achieve mutual objectives. The interest of the private sector in the potential benefits of the program is reflected in a level of cost sharing significantly higher than the required 50 percent (i.e., 63 percent). The technologies being developed are capable of being responsive to the demands of the utility sector for retrofit as well as repowering solutions and key components of industry have used the opportunity to address major pollution issues that threaten their continued existence from an economic and environmental point of view.

The retrofit technologies in the program include six innovative processes for the reduction of sulfur dioxide, six processes for the control of nitrous oxides and six new concepts for their simultaneous removal. These processes introduce combustion modification techniques, new as well as state-of-the-art absorbents employed in new applications and the development and use of new generations of catalysts.

The potential for increased power output is combined with emissions control in the suite of seven repowering technologies being developed. Such promising concepts as pressurized fluidized-bed combustion, coal gasification, circulating fluidized-bed combustion, both atmospheric and pressurized, will be operated as part of a number of innovative integrated power generation systems. These systems offer the potential of increasing the power output of a facility from 50-150 percent, while achieving significant reduction in SO_2 and NO_x emissions.

The three advanced combustor projects are generating new economic systems for coal utilization in both the utility and industrial sectors and the advanced pollution control processes being developed for the steel and the cement industry offer significant and perhaps essential economic solutions to major environmental obstacles.

Finally, a number of projects are introducing the possibility of fuel flexibility through the conversion of coal. While the conversion of coal into other fuel forms is not a new concept, the introduction of environmental issues in conjunction with economic considerations creates a challenge that is being addressed by three of the projects in the Clean Coal Program.

These projects represent options to the utility and industrial user that can accommodate a wide range of site specific requirements to become the solution to emissions control and power production problems.

The role that the technologies play in the reduction and control of acid rain precursor emissions is clearly evident. However, the significant role that some of the technologies can play in resolving some of the complex issues of global warming is not so evident.

Global warming is a critical environmental issue gaining international attention. It focuses on the possibility of significant changes in global climate as a consequence of changes in atmospheric concentrations of "greenhouse" gases--most notably carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), and chlorofluorocarbons (CFCs).

The question of whether the earth is really warming because of the generation of these greenhouse gases is still a matter of considerable debate and scientific analyses. Moreover, there are significant questions about the relative contributions of various human activities. Nevertheless, if scientific evidence ultimately shows that remedial actions are necessary to mitigate climate change, technology innovation and development is an alternative strategy to regulatory constraints.

Some of the projects now in the CCT Program are developing technologies that can cut the emission of "greenhouse" gases in two fundamental ways. First, many of the CCTs improve the efficiency of the conversion of coal to useful energy. Technologies such as pressurized fluidized-bed combustors, integrated gasification combined-cycle systems, and fuel cells will consume less coal per unit of useful energy produced and thus reduce CO_2 emissions. Second, low- NO_x burners, selective catalytic reduction, and other technologies will reduce NO_x emissions, which should reduce N_2O formation.

As global warming becomes an increasingly important issue, the reduction of emission of greenhouse gases will increase in priority, and the worldwide commercial deployment of CCTs will take on even greater significance.

AN APPROPRIATE RESPONSE

The success achieved thus far in the CCT Program as represented by the number of projects, the degree of industrial participation, and range of technologies included, certainly argues well for the potential of the technology development effort and the possibility of it achieving all of its technical objectives.

The 35 projects that are now underway are equally as significant for the promise they offer in generating solutions to a number of the complex interrelated issues of energy, economics, and the environment. These projects and others to follow permit the continued use of coal as an energy source while:

- Being responsive to the requirements of the Clean Air Act Amendments.
- Reducing the amount of acid rain precursors generated during combustion.
- Improving the ability of the utility industry to meet increased demands for power in a cost effective manner.
- Contributing significantly to improved control over carbon dioxide emissions and thus addressing the issue of global warming.
- Providing environmental control options to accommodate a wide range of specific site requirements.

These capabilities certainly indicate that the CCT Demonstration Program can provide appropriate responses to a wide range of complex environmental issues.

THE DILEMMA OF FOSSIL FUEL USE AND GLOBAL CLIMATE CHANGE

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THE DILEMMA

The energy systems of society are both parts of the means to achieve sustainability and the potential causes of instability. Fossil fuels (petroleum, natural gas, coal, oil shale, etc.) epitomize this dilemma. These are our principal energy sources, yet they are depletable on a time scale that is relevant to human history (~200–1000 years), and although their use may be changing the environment of the planet locally, regionally, and even globally (e.g., changing the greenhouse effect), we live in a developing society that demands more energy for more people. The challenge is to avoid the dilemma by technology and policy intervention so that fossil fuels are used to the net benefit of society and its environment.

Of course, concern about the changing greenhouse effect may ultimately limit the use of fossil fuels, and the issue is fiercely debated (Abelson 1990) because uncertainties permeate the entire matter. Although the increase in the concentrations of greenhouse gases in the atmosphere is indisputable, the evidence of consequential temperature or other climate change is not. Still, we tend to agree with T. A. Sanction (1989) that "it is far too risky to do nothing while awaiting absolute proof of the disaster" and with Senator Albert Gore of Tennessee (1989) that uncertainties about the greenhouse effect and the dire nature of the ecological crisis we face should not be used as excuses for inaction.

We should take those low-cost measures that slow greenhouse gas emissions, and we should be prepared technologically to accomplish much larger reductions if necessary. At present, our technological insurance is not in place, but the opportunities for improvement are great, even for fossil fuels.

WHY ARE FOSSIL FUELS SO POPULAR?

Presently, fossil fuels account for about 88% of the commercial energy sources used (not counting energy supplied directly by the sun and traditional biomass sources not traded in commerce). This situation hasn't changed much over the last 50 years (Table 1) and could persist for 50 more. Considering the environmental problems associated with the increasing use of fossil fuels, why are they so popular?

Fossil fuels are relatively marvelous energy sources. The variety of fossil fuels plus the technology mankind has developed to produce and convert them to useful purposes is a marvelous combination.

Furthermore, as a consequence of biomass production during past geologic epochs, when the planet was apparently much warmer, the reservoirs of fossil fuels were built rather ubiquitously. As a result, fossil fuels are available everywhere, and some (e.g., petroleum) are readily transportable. Technical advances have led not only to discoveries and production from the most inhospitable places but also to more complete resource recovery.

Although fossil fuels are depletable, the estimated resources are still very large (Fig. 1). In this figure, reserves are the discovered quantities in known reservoirs and locations that are technically and economically recoverable using current extraction technology. The undiscovered resources of oil and gas are judgmental estimates of those resources thought to be geologically possible and technically recoverable within a reasonable price range. For coal, ultimately recoverable geological resources is an estimate based on the assumption that 50% of the total coal resources-in-place can be recovered using current mining techniques as well as advanced techniques yet to be developed.

Coal, the most abundant fossil fuel, is located predominantly in the U.S.S.R., the United States, and China. Total world resources of coal are estimated to be over 10,000 Gt, and ultimately recoverable resources are estimated to be about 5,500 Gt, or about 150,000 quads (quadrillion Btu). At present use rates, these resources would last 1,500 years.

Oil resources are much less abundant. At historical recovery rates of about 34%, the remaining recoverable resources of conventional oil are estimated to be about 7,000 quads (Masters 1987); which would last only about 60 years at present use rates. But with enhanced oil recovery and the use of unconventional oils, the recoverable oil resources might be doubled.

The remaining recoverable resources of natural gas are distributed rather ubiquitously, but the U.S.S.R. has more than any other region (Dreyfus 1989). Estimates are similar to those of petroleum (about 8,000

quads), which is approximately 120 years supply at current use rates. The ultimate supply might be double that if unconventional sources such as Devonian shales; tight, deep formations; and coal seam gas are considered.

This resource situation would be much more limiting if it were not for the fact that one form of fossil fuel can be chemically transformed into another; for example, coal can be converted to gaseous or liquid fuels. Also, natural gas can be catalytically reformed to produce liquids for transportation albeit at some cost and thermodynamic penalty. The continuing challenge is to develop efficient and economic processes for performing these chemical conversions. Of course, fossil fuels, particularly petroleum and natural gas, are excellent feedstocks for making useful chemicals and plastics.

Fossil fuels are attractive not only because they are available and relatively inexpensive but also because we have learned to use them so effectively. The relatively simple technology of controlled combustion provides energy for both small- and large-scale applications. Almost exclusively, liquids refined from petroleum power the world's transportation systems (greater than 97% in the United States) because these fuels have such a high energy density, because they are so portable, and because of the development of the internal combustion engine and the modern jet engine.

Although many nonfossil energy sources exist, none, either separately or collectively, are ready to substitute for fossil fuels worldwide at the necessary large scale and with the performance, cost, and social acceptance required to be competitive. Nuclear power is perhaps the nearest to being ready, but a significantly expanded deployment is constrained by concerns over reactor safety, accidental reactor damage, and diversion of nuclear fuel to weapons; by problems with managing waste; and by escalating capital and operating costs. Even France, which produces 70% of its electricity by nuclear power, still uses fossil fuels to provide most of its energy (65%). Biomass and hydropower are resource-limited in many countries. Solar thermal electric, photovoltaics, and wind are still expensive, and the power they provide is intermittent. Geothermal sources are geographically constrained and often expensive to develop, as are ocean thermal, wave, and tidal power. Fusion is considered decades away from practical demonstration.

The environmental problems with fossil fuels that command most of our attention today include acid deposition, urban air pollution, and climate change (global warming or the changing greenhouse effect). The acid deposition problem can be solved over time at reasonable costs. In the United States, all urban air quality probably cannot be brought into compliance with all present standards at reasonable cost, but the problem can be kept within acceptable limits (Russell 1988). However, climate change is a different type of problem for which no technological fix yet exists, and the global consequences could be very serious, if not disastrous.

CONTROLLING CO₂ EMISSIONS

Global warming may occur as a result of the release of the so-called greenhouse gases, notably carbon dioxide (CO₂), methane (CH₄), chlorofluorocarbons (CFCs) (e.g., refrigerant gases such as the Freons), nitrous oxide (N₂O), and O₃ (Smith 1988). Because they are relatively long-lived in the atmosphere, dispersion of these gases is much broader than the acid gases, and, because of this wider dispersion, the concern is truly global as opposed to regional. These gases absorb heat energy (infrared radiation) that would otherwise be radiated from the earth to space, resulting in a warming of the troposphere (lower atmosphere). Of these anthropogenic gases, CO₂ is the major one, presently accounting for about one-half of the changing greenhouse phenomenon, and the burning of fossil fuels is estimated to contribute more than 75% of the increasing CO₂ concentration in the atmosphere. The other major source of CO₂ is from deforestation by slash and burn techniques. The consequences of global warming are poorly understood and are not yet predictable in detail, but they could include a 1.5 to 4°C increase in global annual mean-surface temperature for each doubling of CO₂ concentration; marked changes in the amount and distribution of precipitation; large seasonal changes in mean soil moisture; and reduction of some of the world's great ice masses and thermal expansion of the oceans, which would raise sea levels and flood coastal areas.

Our principal concern is how to control greenhouse gas emissions, particularly CO₂. Depending on the fraction of CO₂ retained in the atmosphere, burning all fossil fuel resources could quadruple the CO₂ concentrations in the atmosphere from the present value of about 350 to 1500 ppmv (parts per million by volume) (Table 2). If current models of warming are correct, such an increase in CO₂ concentration would lead to a global average temperature rise in the range of 3 to 8°C with even higher values at the higher northern latitudes.

If such an increase were to occur over a period of two centuries or so, it would likely be both too much and too fast (in the range of 0.15 to 0.4°C per decade). There exists, however, a CO₂ emission rate at which the atmospheric concentration does not increase, or at least it increases very slowly. A carbon cycle model has been used (Emanuel 1990) to examine several scenarios in which the emission rate is suddenly and dramatically reduced from what it is today and then maintained constant at that reduced rate (Fig. 2). The results indicate that CO₂ emission rates must be very low to prevent any increase in CO₂ concentration (of the order of 1 Gt(C)/year); however, rates of 2 to 3 Gt(C)/year lead to only moderate increases over the next 100 years.

Some have argued that the problem is not warming per se but rather the rate of change. A rate of change $<0.1^{\circ}\text{C}$ per decade has been suggested to be slow enough to be manageable. If we assume that half the increase is due to other greenhouse gases, this rate of change translates into an allowable CO_2 emission rate from fossil fuels of 1.6 to 3.5 $\text{Gt}(\text{C})/\text{year}$ where the range depends on the value of temperature change assumed for CO_2 doubling. This emission rate range may be extended upward if emissions of other greenhouse gases are also controlled.

The technological and social management challenge is to get maximum energy services from fossil fuels, hold the emission rate as low as practical, and control the rate within an acceptable range. The challenge is definitely formidable if not impossible. If it becomes necessary to reduce CO_2 emissions to a level much less than the present one, it will take decades to accomplish, and the rate will undoubtedly increase significantly before any reduction can be managed.

Also, the idea that there is some CO_2 emission ration raises allocation issues. Who gets to use what? Does the ration go to countries that have below average fossil fuel consumption? Do industrialized nations reduce their use rates so that the developing nations could use more fossil fuels to spur their economic growth? Most of the greenhouse gases are being generated in the developed world that comprises only a fraction of the world population. Of the two most prominent greenhouse gases, only 25% of CO_2 emissions and $<10\%$ percent of CFC emissions come from the developing world. On a per capita basis, the developing countries have extremely low absolute levels of energy usage and CO_2 emissions (See Figs. 3 & 4).

Indeed, the emission rates of CO_2 have moderated over the past decade and a half as a result of the Arab oil embargo and subsequent oil price shocks. Controlling emissions of CO_2 may be very expensive. A recent calculation (Manne and Richels 1990) indicates that with current technologies and with a ban on nuclear expansion the tax on a ton of carbon emissions would need to go as high as \$600 to force United States emissions to be reduced to 80% of the current emissions of 1.4 $\text{Gt}(\text{C})/\text{year}$, and the cost to the United States economy might be 5% of the Gross National Product. With advanced technologies, this cost might be reduced substantially, perhaps by a factor of 5 or more (Williams 1990).

More Efficient Use of Fossil Fuels. Because nonfossil energy sources are currently poor competitors, it has been argued that improving the efficiency of fossil fuel use is the least expensive path to reducing CO_2 emissions (Keepin and Katz 1988). Indeed, the technological opportunities are very large for all segments of the economy. The major attractiveness of this option is that not only do technologies exist, but their increased applications are often economical even at current fuel prices. In addition, the potential seems to be large for developing nations as well as industrialized nations. This conclusion is brilliantly argued by Goldemberg et al. (1988). These authors conclude that the developing nations can achieve a level of affluence equivalent to that of Western Europe in the mid-1970s by a rate of energy use of only about 1.3 kW/person . This compares to the current level actually used by Western Europe, which is about 4.1 kW/person . In other words, it may be possible for developing nations to grow economically along a much more efficient path than those followed by industrialized nations. Despite the attractiveness of such a high efficiency path, it may prove difficult to achieve the degree that is economically justified without significant activity by governments to encourage it. Many barriers must be overcome, not the least of which are the tendency to make investments based on least first costs rather than least life-cycle costs or the tendency for governments to subsidize energy prices.

As more efficient and economical technologies for fossil fuel use are developed and adopted, however, the more difficult it will be for nonfossil sources of energy to compete. The higher efficiency will mean less CO_2 emissions per unit of energy service, but the reduction will be much less than could be achieved if nonfossil sources were substituted. As always, fossil fuel technology gets better and better, and it is a moving target for its competitors. Greater emphasis must be placed on research and development to improve the nonfossil sources.

Substituting Natural Gas for Coal. The second option for reducing CO_2 emissions from fossil fuels is to substitute natural gas for coal (see Figs. 5 & 6). The heat of combustion per molecule of CO_2 produced is 70% greater than for coal, and natural gas can generally be used more efficiently to produce the same energy services. One reason for higher efficiencies using natural gas is that it does not have the sulfur and nitrogen fuel-bound contaminants and ash content that plague coal and result in acid gas and particulate emissions that must be controlled.

Some of the same repowering technologies that are promising for reducing acid gas emissions from coal-fired electricity generation are also important in reducing CO_2 emissions by substituting natural gas for coal. These include advanced gas turbines and fuel cells; the former is much more advanced than the latter, and further improvements in gas turbine technologies are likely, particularly improvements in materials that will permit higher combustion temperatures and pressures. Additionally, Williams (1989) has proposed catalytic chemical reforming of natural gas with steam to produce hydrogen and carbon monoxide which are then burned in an intercooled steam-injected gas turbine (ISTIG). This chemically recuperated ISTIG may further increase power output, and efficiency may be as high as 52.5%.

Although much less developed than gas turbines, fuel cells offer promise for increasing the efficiency of electricity generation from fossil fuels. Fuel cells are devices (like batteries) that convert chemical energy

into electrical energy. The fuel is oxidized at the anode to provide electrons that flow in the external power circuit to the cathode where oxygen is reduced. The anode and cathode are separated by an electrolyte that provides a transport mechanism for ions but not for electrons.

There are two limitations with this natural gas substitution strategy. First, the resources of natural gas are much smaller than those of coal (Fig. 1). However, substitution of natural gas for coal can be an important interim strategy to moderate CO₂ emissions while better nonfossil sources are developed and deployed. Second, leakage of natural gas from production and transport systems may partially offset the advantage of its use. Methane, the principal constituent of natural gas, is a much more effective greenhouse gas than CO₂. The infrared absorption of a methane molecule is almost 30 times that of a CO₂ molecule. However, the effective lifetime of methane in the atmosphere is much shorter. This problem requires much more investigation because the sources and sinks of methane are not well understood, but it should be possible to reduce leakages from the natural gas system to a negligible value.

The slow pace of growth in natural gas use in the developing world has been largely due to the relatively high cost of transporting the gas from the point of production to the point of use in the era of low oil prices. Indeed, historically, there has been little systematic exploration for gas in these regions, and most of the gas reserves were discovered while looking for oil. However, with the generally higher oil prices prevailing since the 1970s, natural gas use has become economic even in markets far from reserves. As a result, natural gas consumption has been rising, especially outside the United States.

Most gas is still consumed in the country where it is produced, but world trade in natural gas has been rising since the early 1970s. Currently, world trade via pipeline accounts for about 11% of total use and liquefied natural gas (LNG) trade accounts for about 3%. The distribution network in the United States is the most extensive in the world, and it is already linked to supplies from Canada by pipeline and from North Africa by LNG. A potentially useful pipeline link to Mexico also exists and projects are being considered to bring additional LNG from Trinidad, Venezuela, and Nigeria to the United States.

The gas distribution network in Western and Eastern Europe is also well developed and is linked by pipeline to large producing fields in The Netherlands, Norway, the U.S.S.R., and North Africa. Countries that are not already linked to the grid (Greece, Portugal, Turkey, etc.) are making plans to join soon. Recently, Iran has reactivated its earlier plan to supply Western and Eastern European countries by pipeline through the Soviet Union. Some LNG is coming into Western Europe from Algeria and Libya currently, and plans are being made to bring LNG from Nigeria.

While the major Eastern European countries have already integrated some natural gas from the Soviet Union into their energy mix, they still face immense environmental problems because of their heavy reliance on coal. These countries offer a very good opportunity for replacing coal with Soviet and Middle Eastern natural gas.

Japan has made considerable progress in natural gas usage with LNG imported from Indonesia, Malaysia, Australia, Alaska, and the Middle East, but consumption elsewhere in Asia is relatively modest, based mostly on available indigenous resources. However, the picture is changing. Korea has recently started LNG imports, and Taiwan will soon begin. A proposal for an Asian grid linking the producing fields in Indonesia, Malaysia, and Thailand with other Pacific Rim countries is also being pursued, and may be implemented during the 1990s (Cedigaz News Report 1990). India is exploring possibilities of importing LNG from the Middle East to supplement its own rising production. Iran is promoting a \$12-billion project to bring its prolific natural gas resources to India and Pakistan by a 2000-mile pipeline from Bandar Abbas to Calcutta (Petroleum Economist 1990). The Soviet Union has recently initiated discussions with Japan to build a 3,100-mile pipeline, partly undersea between Siberia and Japan via South and North Korea (Wall Street Journal 1990).

International trade within South America is still limited, but recent bilateral agreements between Argentina, Bolivia, and Brazil portend expansion.

Helping the Soviet Union with advanced gas production, transmission, and utilization technologies would seem to be a stabilizing policy for the OECD countries to adopt in this day of "perestroika." Expanding the use of Middle East gas could make an important contribution, not only to Europe but also, perhaps, to Pakistan and India as well. Resources of Indonesia and Malaysia will likely underpin the growth of natural gas consumption in the Pacific Rim nations. Helping these countries use gas resources is environmentally sound as an interim strategy, and also economically desirable as a developmental strategy. These steps could have a very positive impact on the economics of the Soviet Union, the developing regions of the world, and on urban and regional ambient air environmental quality.

Recovering and Sequestering CO₂. The third strategy for reducing CO₂ emissions from fossil fuel is to capture the emissions and sequester them or find nondispersive uses for the recovered CO₂. This is an expensive proposition, at least for the techniques suggested so far. The difficulty of the problem is evident by the simple realization that 1 ton of carbon in fossil fuel produces almost 4 tons of CO₂.

One technique, of course, is to grow forests and recycle the carbon back into the biomass reservoir. Recently, Advanced Energy Services agreed to fund planting trees in Guatemala sufficient to offset the CO₂ put into the atmosphere during the lifetime of a proposed new coal-fired plant in Connecticut (Pearce 1988).

Although this is an interesting and perhaps important approach to afforestation, it probably cannot be practiced at the scale that would be required to make a significant offset to CO₂ released worldwide from fossil fuel use. For example, offsetting the emissions of a 500 MWe coal fired power plant operating at about 34% efficiency would require about 500 square miles of forest to be grown assuming the forest fixes 2 tons of carbon per acre per year over the lifetime of the coal plant. To sequester the total carbon emissions of the United States would require growing about one million square miles of forests at this productivity and using about 25% of the land area of the United States. Growing trees could, however, do much to offset the deforestation trend.

It should be acknowledged that the role of biomass in the global carbon cycle is still largely unknown. For example, Tans, Fung, and Takahashi (1990) recently analyzed the northern-hemisphere-southern-hemisphere CO₂ gradients and concluded that there must be a large northern hemisphere sink, presumably of terrestrial origin. If the analysis is correct, the effect could be due to CO₂ fertilization causing plants to grow more rapidly or to afforestation in the northern hemisphere. Much more needs to be learned about the natural carbon cycle.

Other strategies have been proposed for recovering CO₂ from the emissions of large fossil fuel facilities such as power plants. These involve recovering CO₂ exhaust gases and permanently sequestering the CO₂ either in the deep oceans or in depleted natural gas reservoirs or other geologic formations such as the hollowed-out salt domes used in the United States for storing strategic petroleum reserves. Steinberg (1985; Horn and Steinberg 1982) was one of the first to look at these possibilities carefully. He estimated that the added cost of recovering and sequestering CO₂ emissions would increase the cost of electricity generation by as much as a factor of 1.8 to 5. However, a recent calculation revises this range substantially downward (Table 3).

In summary, we suspect that CO₂ sequestering schemes (except perhaps for reforestation which, as noted, has its own limitations) will always prove to be more expensive than substituting nonfossil sources, but the calculations of Hendriks, Blok and Turkenburg (1990) clearly narrow the gap and point to needed research.

Hydrogen From Fossil Fuels. Fossil fuels can also be used as a source of hydrogen, an alternative to hydrocarbons as a fuel material. Carbon, hydrocarbons, and carbon monoxide are splendid reducing agents for producing hydrogen from water. Of course, a product of the reduction of water by carbonaceous materials is still CO₂, which must be recovered and sequestered. Nevertheless, we could imagine the use of hydrogen derived from fossil fuels as a clean source of energy for Los Angeles. In fact, it is not inconceivable that it could be an interim step to a situation 50-100 years hence when the principal energy carriers used by society are electricity, hydrogen and biomass-derived liquids for transportation.

To this end, Williams (1990) has used the calculations of Hendriks, Blok and Turkenburg (1990) to estimate the cost of producing hydrogen from coal using the Shell oxygen blown gasifier and sequestering the CO₂ in depleted gas or oil reservoirs. The estimates range from \$4.50 to \$5.74/million Btu on the basis of higher heating value. The lower cost assumes a 6% discount rate whereas the higher cost assumes a 12% discount rate. This is an extremely interesting range of costs given the potential importance of hydrogen as a clean fuel.

By adding heat from nonfossil sources such as from a high-temperature gas-cooled nuclear reactor or perhaps a solar furnace, the yield of hydrogen per unit of CO₂ produced can be increased. For reforming of methane, hydrogen production can be increased by 1/3 and by 70% from coal. Whether such heat sources to boost hydrogen production from fossil fuels will be practical depends on the cost of competing sources of hydrogen such as direct electrolysis of water using nuclear, solar, or wind electricity.

Hydrogen could be produced near coal fields, the CO₂ pumped to sequestering sites, and the hydrogen piped to centers of fuel use. However, hydrogen is not very portable. Gaseous hydrogen at 2400 psi has a volume about 18 times the volume of an equivalent amount of energy stored as gasoline and weighs three times as much if the weight of the storage vessel is included. Hydrogen can be stored as metal hydrides and released by heating. Hydrides weigh about as much as high-pressure gas cylinders but occupy only about 25% more space than a gasoline tank containing the same energy value (Amann 1990). By increasing the efficiency of conversion dramatically, these storage limits may one day be acceptable even for automotive transportation systems. As we have mentioned, hydrogen can be used very efficiently; for example, by fuel cells where conversion of chemical energy to electricity may be accomplished at 60% efficiency, indicating the possibility of a practical hydrogen fuel cell electric vehicle.

From this discussion it is clear that the near-term moderation of the emissions of CO₂ from fossil fuel use depends on two strategies. The first is to use fossil fuels more efficiently across the board from conversion to end uses. This applies both to the conversion to electricity and the efficient use of electricity. Of course, the development and adoption of much more efficient technologies for using petroleum products particularly for transportation is an essential ingredient. The second, related to the first, is to substitute high efficiency gas technologies for coal wherever practical. Stimulation of the use of natural gas at the expense of coal will, of course, increase natural gas prices and hasten depletion of resources. But it can also buy time to develop better nonfossil sources and improve air quality at the same time. Sequestering of CO₂ emissions from the exhausts of fossil fuel combustion or conversion seems presently expensive and impractical except, of course, as it is accomplished by afforestation. Nevertheless, the possibilities including that of hydrogen production should be intensively researched.

CONCLUSIONS — MANAGING FOSSIL FUELS

Fossil fuels can only make a transient contribution to the energy supply for a sustainable planet because they are finite and depletable resources. Nevertheless, that transient contribution is most significant because fossil fuels are mankind's primary commercial energy source, and their use worldwide is growing, especially by the developing nations. Fossil fuels are so important because they are still relatively abundant, cheap, ubiquitous, and some forms (notably petroleum) are readily transportable. Hydrocarbon liquids with high energy density are portable and make superb fuels for powering transportation systems, and one form of fossil fuel can often be readily converted to another (e.g., gases and solids to liquids and solids to gases). Furthermore, they can be used at almost any scale employing simple or complex technology, and they are the source of an enormous variety of chemicals and plastics.

Nevertheless, as the use of fossil fuels has grown, the problems of protecting the environment and human health and safety have also grown, providing a continuing challenge to technological and managerial innovation. Today that challenge is to control atmospheric emissions from combustion, particularly those emissions that cause acidic deposition, urban pollution, and increasing concentrations of greenhouse gases. Technology for reducing acidic deposition is available and needs only to be adopted, and the remedies for urban pollution are being developed and tested. How effective or expensive these will be remains to be determined. The control of emissions of the greenhouse gas, CO_2 , seems possible only by reducing the total amounts of fossil fuels used worldwide, and by substituting efficient natural gas technologies for coal. Long before physical depletion forces the transition away from fossil fuels, it is at least plausible and even likely that the greenhouse effect will impose a show-stopping constraint. If such a transition were soon to be necessary, the costs would be very high because substitute energy sources are either limited or expensive or undesirable for other reasons. Furthermore, the costs would be unevenly felt and would be more oppressive for developing nations because they would be least able to pay and, on average, their use rates of fossil fuels are growing much faster than those of many industrialized countries.

It is prudent, therefore, to try to manage the use of fossil fuels as if a greenhouse constraint is an important possibility. This suggests taking several low-cost actions in the near-term such as:

- policies including R&D that encourage the development of more efficient and economical end-use and conversion technologies (e.g., more efficient gas turbines, fuel cells, oxygen-blown gasifiers, and processes for producing hydrogen from fossil sources);
- more intensive R&D to accelerate the development of better nonfossil sources including direct solar, biomass and other renewables, fission, and fusion;
- policies that encourage the substitution of hydrogen-rich for hydrogen-lean fuels and expansion of the natural gas system, particularly in developing nations;
- cooperation by western industrialized countries in providing technical assistance to developing and Eastern Bloc countries for producing energy technologies that are both economically and environmentally more attractive, including expanding the development and use of the natural gas resources of the Soviet Union;
- R&D to increase our understanding of the global cycle of CO_2 releases to the atmosphere and its removal by the oceans and by terrestrial ecosystems so that climate stabilization targets and policies can be better established; and
- experiments with CO_2 recovery and sequestration techniques so that the economic and environmental impacts are better understood.

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Table 1. World primary energy use and associated CO₂ emissions by year for the past 50 years

	Oil		Gas		Coal		Fossil		Hydroelectric		Nuclear		Total	CO ₂	E/P
	q	%	q	%	q	%	q	%	q	%	q	%	q	Gt(C)/y	MMBtu
1937															
US	7	33	2	11	12	55	21	99	0	1			21	0.4	161
World	12	20	3	5	45	75	60	99	0	1			60	1.2	29
1947															
US	12	36	4	13	16	50	32	99	0	1			32	0.7	221
World	18	24	6	9	48	66	72	99	0	1			72	1.4	32
1957															
US	18	44	10	25	11	27	39	96	2	4			40	0.8	236
World	35	33	12	11	53	50	100	94	6	6			106	2.2	37
1967															
US	25	44	18	31	12	21	55	96	2	4			58	1.0	290
World	70	40	30	17	65	37	165	94	11	6			176	3.3	51
1973															
US	35	47	23	30	13	17	70	95	3	4	1	1	74	1.3	351
World	111	47	42	18	66	28	220	94	13	6	2	1	235	4.5	60
1977															
US	37	49	20	26	14	18	71	93	3	3	3	4	76	1.3	346
World	118	46	46	18	73	28	237	92	15	6	5	2	258	4.8	61
1985															
US	31	42	18	24	17	24	66	90	3	5	4	6	74	1.3	309
World	112	38	59	20	90	31	260	88	20	7	14	5	295	5.3	61
1986															
US	32	43	17	23	17	23	66	89	3	5	4	6	74	1.3	308
World	115	38	59	20	92	31	266	88	21	7	15	5	302	5.4	61
1987															
US	33	43	17	23	18	24	68	89	3	4	5	6	76	1.3	312
World	117	38	62	20	95	31	273	88	21	7	16	5	310	5.5	62
1988															
US	34	43	18	23	19	24	71	89	3	4	6	7	80	1.4	
World	121	38	65	20	96	30	282	88	22	7	17	5	320	5.5	

q - quads (quadrillion Btu); Gt(C) - gigaton (billion metric tons) carbon as CO₂; y - year;
E/P - Energy per capita (person); MMBtu - million Btu

Although alternative energy sources, most notably nuclear and hydropower, have become available during the last fifty years, total energy production from fossil energy sources has grown dramatically. Percentagewise, fossil fuels will probably continue to shoulder much of the energy burden for many years to come because alternative energy sources are either not economically competitive or cannot be implemented on a large scale.

[Source: Fulkerson, 1989]

Table 2. If all recoverable resources of fossil fuels were burned, significant increases in atmospheric CO₂ would result, with the absolute magnitude of the increases being dependent on the fraction of CO₂ released that is retained in the atmosphere. Of the fossil fuels, only coal is sufficiently abundant to increase atmospheric CO₂ by more than a factor of two.

Fuel	Recoverable Quantity	Energy Value (1000s of Quads)	Carbon ^a Content 10 ¹⁵ g	CO ₂ Concentration Increase (ppm) ^b Fraction Retained in Atmosphere		
				0.4	0.55	0.7
Oil	1255 x 10 ⁹ bbl	7	130	24	34	43
Gas	8200 tcf	8	120	23	31	39
Coal	5500 x 10 ¹⁵ g	<u>153</u>	<u>3850</u>	<u>723</u>	<u>994</u>	<u>1265</u>
Totals	(Rounded off)	168	4100	770	1060	1350

Source [Fulkerson, et al., 1989]

^aIn addition to these amounts of carbon, comparable or larger amounts may be available in other fossil resources such as heavy oils, oil shales, tar sands, lower grades of coal, etc. Thus, the quantity of carbon ultimately released to the atmosphere as CO₂ could conceivably be half again as much, or twice as much, as the total shown in the table.

^bThese hypothetical increases may be compared with the preindustrial CO₂ concentration (about 270 ppmv), the present concentration (350 ppmv), or the current annual increase (about 1.5 ppmv/year). In the atmosphere, 1 ppm of CO₂ by volume, uniformly distributed, equals about 2.13 Gt of carbon, or 7.81 Gt of CO₂. Thus, 350 ppmv CO₂ corresponds to 745 Gt C. (1 Gt = 10⁹ metric tons = 10¹⁵ g)

Table 3. The estimated costs of CO₂ recovery and sequestration from central power plants are large, but may not be prohibitive. The economically viable method at present appears to be afforestation, but it is severely limited.

Removal Process	Relative Increase In Electricity Cost	Reduction in Efficiency for CO ₂ removal	% CO ₂ removed
Lime adsorption	2.4 ^a	59	90
Monoethanolamine Scrubbing	2.3 ^a	57	90
O ₂ Combustion	1.8 ^a	30	100
IGCC/Selexol (Oxygen blown gasifier plus air combustor)	1.3 ^b	13	88
Afforestation	1.1	0	100

a--Includes deep ocean disposal

b--Includes disposal in depleted natural gas wells

Sources: Steinberg, 1985, 1986, 1987; Blok, Hendrix, and Turkenburg 1989;
Hendrix, Blok and Turkenburg, 1990; Golomb et al. 1989.

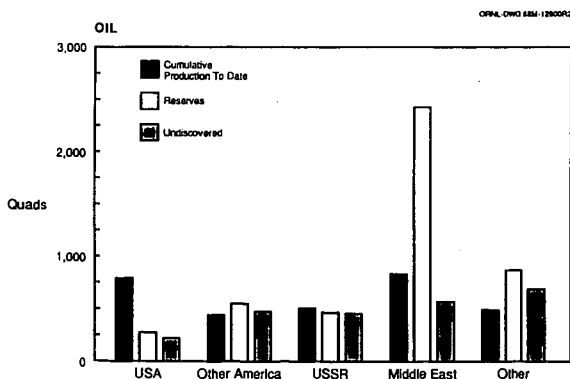
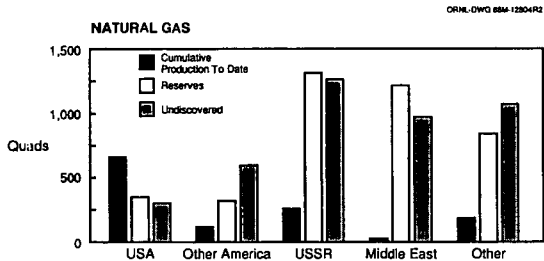
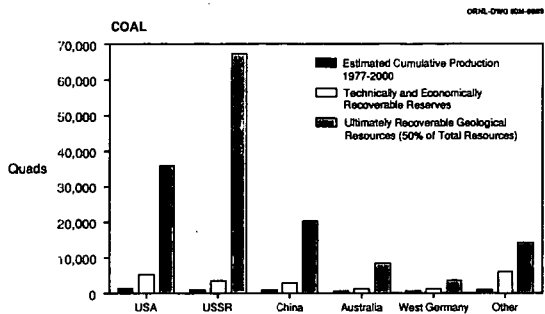


Figure 1. Fossil fuels are presently our most used and obtainable energy resources. They are dispersed throughout the world, and coal is by far the most abundant of the fossil fuels. Although coal is depletable, it would last about 1500 years at present use rates; oil, 60 years; and gas, 120 years.

Sources: Masters, et al., 1987; Wilson, 1980.

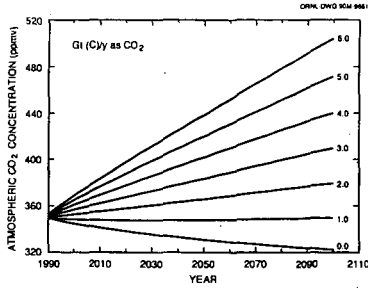


Figure 2. Atmospheric concentrations of CO₂ calculated by assuming that the current rate of emissions is suddenly changed to the values indicated and maintained constant thereafter.

Source: Emanuel 1990.

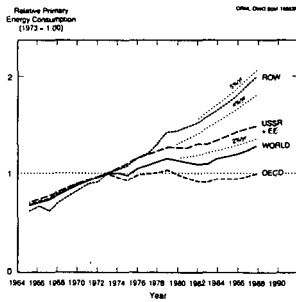


Figure 3. Relative world consumption of fossil energy 1965-1988, by regions.

Source: BP 1989.

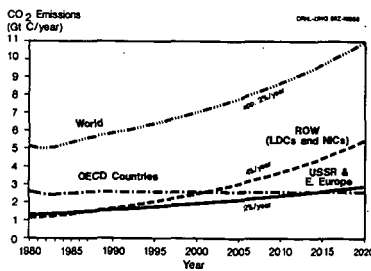


Figure 4. Extrapolation of CO₂ emission growth rates for the decade 1977 to 1987.

Source: Fulkerson 1989.

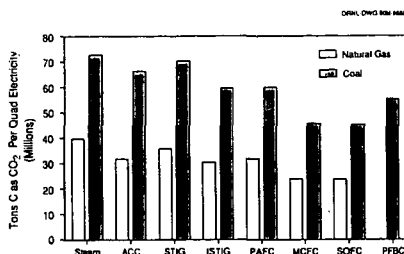


Figure 5. Carbon emissions from electricity generation using fossil fuels depend in large measure on the fuels used and the efficiencies of the generation techniques used. This figure compares emissions of carbon, expressed in terms of millions of tons of carbon as CO₂ per quad of electricity produced, for several electricity generating options using natural gas and coal as fuels. The options include conventional steam-electric plants (Steam), advanced combined cycle (ACC), steam-injected gas turbines (STIG), intercooled steam-injected gas turbines (ISTIG), phosphoric acid fuel cells (PAFC), molten carbonate fuel cells (MCFC), solid oxide fuel cells (SOFC), and pressurized fluidized-bed combustors (PFBC).

[Millions of metric tons C as CO₂ emitted per quad of electricity = 52.5 (natural gas) or 92.0 (coal) x (12/44)/efficiency; for example, for a steam plant fired with an efficiency of 34.6%, the amount of carbon dioxide that is released per quad is $92 \times 0.2727/0.346 = 72.5$ million tons]

Sources: *Clean Coal Technology*, 1989; Pillai, 1989; Schora and Camara, 1990; Williams and Larson, 1989; Blomen, 1989.

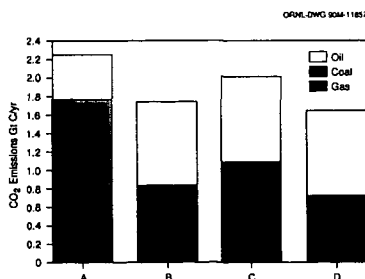


Figure 6. Four scenarios for CO₂ emissions from the use of fossil fuel by the USSR, other Eastern European countries, and Western European countries. Scenario A assumes future emissions are the same as for 1988. Scenario B assumes all coal use in Scenario A is replaced by natural gas but with efficiencies 20% greater than coal. Scenario C is the same as Scenario B except that all oil use is replaced by methanol made from natural gas at a conversion efficiency of 60%. Scenario D is the same as Scenario C except that nuclear or solar heat improves the efficiency of conversion to 90% and the CO₂ emissions are reduced accordingly. In Scenario A, 34 quads of natural gas are used per year; in Scenario B, 645 quads; in Scenario C, 141 quads; and in Scenario D, 115 quads. The estimated recoverable gas resources and reserves for Europe would last about 90 years for Scenario A, but only 45 years for Scenario B and 21 and 26 years for Scenarios C and D, respectively.

Diversification of Raw Materials for
Domestically Produced Transportation Fuels

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Keywords: Transportation Fuels, Synthetic Fuels, Alternative Fuels

In the past the transportation fuels, gasoline, jet and diesel fuel, and residual fuels have been almost entirely manufactured from petroleum and natural gas liquids, with growing dependence on foreign sources--currently about 50%.

More recently increasing amounts of oxygenated fuels are blended into gasoline. This change is largely driven by environmental and health concerns. An example is the phasing out of tetraethyl lead which has made limited use of high octane number oxygenated additives cost effective. Urban environmental concerns have resulted in legislation requiring increased gasoline oxygen content. The 1990 clean air act revision requires, for example, 2.7% oxygen content in gasoline by 1992 in some cities during the part of the year when they are prone to high CO levels. At present, oxygenated compounds are based on alcohols from agricultural sources or from natural gas thus providing some resource diversification without major automobile modification. Use of compressed natural gas and methanol fuel, which require vehicle modification, is being introduced for fleet use, largely in urban areas. It is expected that diversification of gasoline composition will continue. Jet fuel and diesel fuel, however, are not expected to undergo major changes.

This paper focuses on the future supply of domestically produced transportation fuels, their cost and the consequent timing for development of fuels from domestic resources. It makes extensive use of information acquired for the National Research Council study "Production Technologies for Liquid Transportation Fuels".⁽¹⁾ In order to focus the discussion, it is assumed that consumption of transportation fuels will remain constant and that we will work toward the goal of maintaining domestic production at 50% of the total.

In the United States, liquid fuel use is dominated by the transportation sector. It is expected that the transportation sector's share of liquid fuels use will grow as petroleum cost increases and supplies diminish since the other sectors can switch relatively easily to natural gas, solid fuels or electricity.

Within the transportation sector the major fuel is motor gasoline. It is not expected that the distribution will change drastically for the next few decades although jet fuel may well continue to grow in importance.

Table 3 presents estimates of remaining economically producible US. crude oil resources as a function of cost and level of technology used. These estimates were prepared for the National Research Council by ICF Industries and take into account the recent increases in resource estimates based on advances in identification of isolated pockets of oil which had previously been overlooked. Tar ($> 10^4$ centipoises) is included in these estimates. "Moderate Cost" is taken as 25 \$/bbl and "High Cost" 40 \$/bbl.

For current practice, which includes secondary recovery, and a cost of \$25/bbl, 75 billion bbl can be recovered. The ratio of this resource to the 1989 production of 3 billion bbl of oil/year provides a time scale for this resource. It is, however, not a prediction of suddenly running out of oil in 25 years. It is expected that, in the near term, production will continue to decrease, but that a vigorous drilling and development program can bring production back to the present rate. In time, production for a given price will decrease as the lower cost resources are consumed. This ratio is, however, useful for planning purposes.

Increasing oil price to \$40/bbl with no change in technology is estimated to increase the economically producible resource to 95 billion bbl (26%). At this price, however, extended oil recovery techniques become economically feasible and 140 billion bbl can be economically produced with a resource to current annual production ratio of 47. These times are the same general magnitude of the time required to build up an industry and facilities for major production of liquid fuel manufactured from alternative resources such as biomass, coal, and oil shale.

In Table 4 estimates for production of domestic natural gas are presented. The wellhead gas price of 3\$/MCF corresponds to an oil price of about 24 \$/bbl. Domestic gas production, on a heating value basis, is currently approximately the same as oil production and the base case resource base/current production ratio is 33 years--32% higher than shown in Table 3 for the petroleum base case. It is expected,, however, the use of natural gas for heating and power generation will grow relative to petroleum (2) so the time scales, may not be greatly different from petroleum. While natural gas can be converted to, or used for, transportation fuel, the domestic resource cannot be expected to substitute for petroleum in transportation fuels to a major extent.

Increasing use of solid fuel resources for manufacture of transportation fuel is indicated by the above considerations. Estimates of the magnitude of these resources are presented in Table 5.

Coal and Western oil shale represent resources large enough to replace domestic petroleum for many years. Estimates of the potential for environmentally and economically acceptable biomass production vary widely (about a factor of on either side) of the estimate shown here. This potential can be compared to the 2.4 billion bbl/yr gasoline shown in Table 2. On this basis, the biomass resource is large enough to be an important source of domestic transportation fuel.

The estimated cost of producing transportation fuels from non-petroleum resources has been estimated using a common basis for process and raw material costs. These estimates are based on a study prepared for the National Research Council by SFA Pacific Inc. ⁽⁴⁾. These costs are expressed as the crude oil cost that would make a spark ignition vehicle fuel from the alternative resource just as expensive to the end user as gasoline from crude oil. The cost estimates are for 1988 dollars with 10% DCF return. Capital costs include vehicle modifications for alternative fuels as well as the investment in manufacturing and distribution facilities.

Natural gas costs, for hydrogen and synthesis gas production, increase with increasing crude oil cost according to the formula

$$\$/\text{MCF} = 3.91 + .05857 (P_{\text{oil}} - 28)$$

P_{oil} is the price of crude oil in \$/bbl.

Heavy oil and tar sands, which were counted as petroleum in Table 3, can be converted to transportation fuels by hydroprocessing, using natural gas as a source of hydrogen, at 25 and 28 \$/bbl equivalent crude cost. Here, as with natural gas, large foreign resources will probably be available at a lower cost for many years.

The use of compressed natural gas for automotive fuel, which includes distribution and vehicle modification costs, was estimated to be 34\$/bbl equivalent crude cost. Equivalent crude costs for coal liquefaction and Western Shale oil were estimated to be 38 and 43 \$/bbl respectively. Over the last decade significant progress has been made in coal liquefaction; however, the shale oil process has suffered from lack of published activity. Leads for further cost reduction, in both processes, offer the potential of achieving, with continued R&D, a cost of 30 \$/bbl for both processes within the next decade.

Methanol by coal gasification is 35-40 \$/bbl. In this case the price of domestic gas makes coal gasification the most economic route. Estimated costs for methanol from overseas natural gas are also shown for U.S. investment costs, but with the cost of transportation to the U.S. added. A 15% energy efficiency advantage and a \$100 automobile cost differential for methanol was also assumed. On this basis, imported methanol based on low cost gas can be lower cost than domestic production from solid fuel resources.

The use of wood for methanol production is one of the more attractive methods for using biomass. In this study wood and coal feed costs were approximately the same and, for the same size of plant, gasification and process costs would be about the same. The costs shown for coal to methanol, however, are based on a large plant (oil equivalent 50,000 bbl/day-oil). Biomass based operations on a smaller scale would have increased costs.

From the above studies it appears that in the 30-40 \$/bbl range of equivalent crude cost, use of all the solid resources becomes of economic interest with heavy oil processing becoming economic at 25 \$/bbl. The time at which prices in this range will become sufficiently stable to attract unsubsidized private capital is unpredictable. If a decision is made to maintain domestic production at a stable level (say 50% of the total), construction of and operation of facilities before the time of stable international oil prices in the 30-40 \$/bbl range will be necessary. Facilities for hydroprocessing of heavy oil and tar are expected to be the first step followed by introduction of use of solid resources. It has been shown that heavy oil hydroprocessing operates successfully with a mixture of coal and oil and that hydrogen, which is used in large quantities can be manufactured from a wide variety of feeds. The possibility exists, therefore, of substantial flexibility in the type of feed used in a given plant with the potential for changing the mix as economic and supply change with time.

These process possibilities are illustrated in Fig. 1. Gasification by partial oxidation is capable of converting any carbonaceous feed to a gaseous mixture from which the hydrogen and CO can be separated. From this hydrogen/carbon monoxide mixture, hydrogen for direct liquefaction can be produced. Methanol or paraffinic liquid fuels, both excellent transportation fuels, can also be manufactured from the hydrogen-carbon monoxide mixture by catalytic conversion.

In direct, or liquefaction, hydrocarbon fuels are produced by reaction of tars or solid fuels with hydrogen produced by gasification.

The production of the greenhouse gas, CO₂, could well have a major effect on the choice of raw materials for transportation fuel. Table 7 presents information on the formation of greenhouse gases, relative to petroleum based gasoline, by production and use of automotive fuels from several starting materials.

Biomass based fuels are considered to be greenhouse gas neutral. This assumes no consumption of fossil fuels in the complete fuel cycle. Methanol from coal, however, produces approximately twice the CO₂ compared to petroleum.

Oil shale is intermediate with the amount of CO₂ produced depending on the degree of carbonate decomposition during the retorting process. Natural gas based fuels are somewhat better than petroleum.

If process heat and hydrogen from non-fossil resources (Biomass, solar, nuclear) becomes available, the greenhouse gas production from gasoline or methanol for all sources is comparable to or better than gasoline from petroleum. Development of non-fossil heat and hydrogen could, therefore, be an important factor in use of our fossil fuel resources.

Conclusions

1. If production of transportation fuels from domestic resources is to be maintained at approximately the current level, it is time to begin the transition from petroleum to alternative raw materials.
2. The 1988 equivalent crude cost, starting with solid resources, is in the 30-40 \$/bbl range.
3. Both supply and environmental considerations lead to a diverse set of raw materials.
4. Co-processing of raw materials offers an opportunity for optimizing the transition to use of solid resources.
5. The production of CO₂ from fossil resources can be substantially reduced by developing non-fossil fuel based sources of heat and hydrogen.

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Table 1

Yearly U.S. Liquid Fuel Use by Sector
(1988)²

<u>Sector</u>	<u>Quads (10¹⁵BTU)</u>	<u>% of Total</u>
Transportation	21.0	72
Residential	1.2	4
Commercial	1.0	3
Industrial	4.8	16
Electric Utilities	1.3	5
	<u>29.3</u>	<u>100</u>

Table 2

Yearly Transportation Fuel Use
(1988)²

<u>Fuel Type</u>	<u>Quads(billion bbl/yr)</u>	<u>% of Total</u>
Motor Gasoline	13.8(2.4)	66
Diesel Fuel	3.5(0.6)	17
Jet Fuel	3.0(0.5)	14
Residual Fuel	0.7(0.1)	3
Total	<u>21.0(3.6)</u>	<u>100</u>

Table 3

Estimated Remaining Economically
Producible US. Crude Oil Resources - 1989⁽³⁾

	<u>Current Practice</u>		<u>Advanced Technology</u>	
	<u>Mod. Cost</u>	<u>High Cost</u>	<u>Mod. Cost</u>	<u>High Cost</u>
Billion bbl oil (QUADS)	75 (435)	95 (551)	115 (667)	140 (810)
Resource base to current annual production ratio	25	32	35	47

Table 4

Estimated Remaining Economically
Producible Natural Gas Resources (3)

	<u>Current Technology</u>		<u>Advanced Technology</u>	
Wellhead Gas Price Dollars/Mcf	3	5	3	5
Tcf Gas (Bbl oil equivalent)	595(107)	770(140)	880(160)	1420(256)
Ratio of Resource Base to Current Production	33	43	50	80

Table 5

Alternative US. Resources (1)

<u>Resource</u>	<u>Recoverable Amount - Quads</u>
Coal	6,000 - 11,000
Western Shale	3,000 - 4,000
Eastern Shale	400
Tar Sands	400
Biomass - Methanol	(~1 billion bbl/yr of oil equivalent)

Table 6

Equivalent Crude Cost of Alternative Fuels
(in 1988 dollars/bbl, at 10% discounted cash flow) (1)

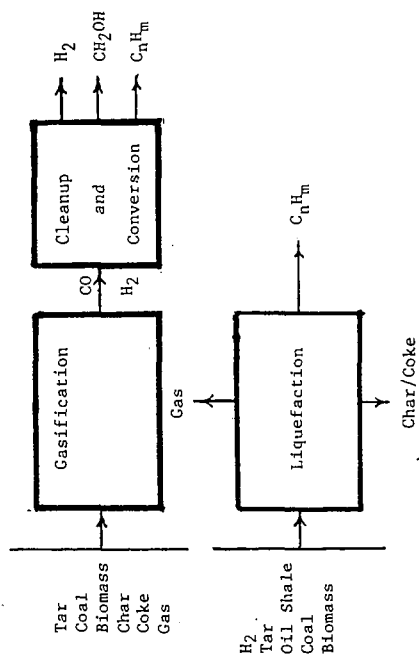
<u>Process</u>	<u>Cost Estimates for Current Published Technology</u>	<u>Cost Targets for Improved Technology</u>
Heavy Oil Conversion	25	-
Tar Sands Extraction	28	-
Coal Liquefaction	38	30
Western Shale Oil	43	30
Compressed Natural Gas	34	-
Methanol via		
Coal Gasification	35-40	-
Natural gas at		
\$4.89/Mcf	45	-
\$3.00/Mcf	37	-
\$1.00/Mcf	24	-

Table 7

Approximate Greenhouse Gas Emission per Mile
Relative to Petroleum-Powered Internal Combustion Engines⁽⁵⁾

Fuel and Feedstock	Percent Change
Current Technology	
Gasoline and diesel from crude oil	0
CNG, gasoline, diesel, or methanol from biomass	-100
Methanol from coal	+ 98
Gasoline from oil shale	+ 27 to + 80
CNG from natural gas	- 19
Methanol from natural gas	- 3
Potential Advanced Technology	
Gasoline from coal or shale using nonfossil sources for process heat and hydrogen	0

Fig. 1
Feeds for Direct and Indirect Solid to Liquid Fuels Conversion



GREENHOUSE GAS EMISSIONS, ABATEMENT AND CONTROL: THE ROLE OF COAL

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Keywords: greenhouse gas, efficiency improvements, CO₂ control technology

ABSTRACT

The basis for quantifying the relative effect of greenhouse gas emissions from coal utilisation is discussed. Emission factors (gC/MJ) need to include greenhouse gas emissions and energy losses throughout the fuel cycle in order to compare the fuels. Nevertheless CO₂ is the most important greenhouse gas from coal. Emission factors have decreased due to improved efficiency of coal use. The scope for further improvements in efficiency of conventional and advanced power generation is assessed. Control of CO₂ emissions is viewed as a less promising option owing to the high cost and energy penalty of most methods.

It is concluded that there is no firm basis for evaluating the effect of reducing emissions on their global warming potential. However it is desirable that available technologies be implemented to reduce emissions by improved efficiency.

INTRODUCTION

Greenhouse gases from coal include methane (CH₄), released during hard coal production, and most important, carbon dioxide (CO₂) as well as nitrous oxide (N₂O) which are emitted when coal is used. Ozone (O₃) is another greenhouse gas formed indirectly from complex reactions with other pollutants in the atmosphere. Here other oxides of nitrogen, nitric oxide and nitrogen dioxide (jointly referred to as NO_x), play a role and are emitted from coal use. The topic of nitrogen oxide emissions from coal combustion was reviewed recently at IEA Coal Research by Sloss (1991). Chlorofluorocarbons (CFC) are important greenhouse gases which are produced in the industrial sector where coal is not involved.

Policy choices over the next few years to reduce greenhouse gas emissions are likely to have a profound effect on the use of coal in many countries. It is therefore essential to examine and monitor the scientific and technical basis for reducing emissions of greenhouse gases from coal use. A major scientific assessment of the greenhouse gas issue is provided by the recent report (Houghton and others, 1990) of Working Group I of the Intergovernmental Panel on Climate Change (IPCC), set up in 1988 by the World Meteorological Organization and the United Nations Environment Programme. The IPCC considered that a reduction in CO₂ emissions >60% would be required to stabilise CO₂ concentrations in the atmosphere. IEA Coal Research has been keeping a watching brief on this issue since 1977 (Smith, 1978, 1982, 1988; Vernon, 1990). This paper examines the basis of the quantitative evaluations which are being commissioned by policy makers in attempting to deal with the greenhouse gas issue and assesses the state-of-the art of abatement and control technologies as they relate to coal use. Information is drawn from a new draft report on this topic (Smith and others, 1991).

GREENHOUSE GAS EMISSIONS

The contributions (%) of trace gases to the greenhouse effect are estimated for the IPCC by Shine and others (1990) over the past decade (based on increased concentrations in the atmosphere) and integrated over the next 100 y (in terms of emissions from human activities in 1990) as follows:

	1980-90	100 y
CO ₂	56	61
CH ₄	15	15
N ₂ O	6	4
CFC	24	12
others	-	8
	101	100

The indirect effects of CH₄ oxidation (the production of CO₂, stratospheric water vapour and

tropospheric O_3) are potentially significant but difficult to quantify. This applies also to the indirect effects from other trace gases: CO (1%), NO_x (6%) and non-methane hydrocarbons (0.5%). Shine and others emphasise that the quantification of these indirect effects needs further revision and evaluation and does not include some important chemical reactions. Victor (1990) points out that the indirect effects vary spatially as well as with time.

Further uncertainties arise from different ways of estimating the effective residence time for CO_2 in the atmosphere. The uptake of CO_2 into the various storage sinks of the global carbon cycle can take a short time, for example into non-woody vegetation or a much longer time into the deep ocean. The IPCC adopted a range of 50–200 y for the effective residence time of CO_2 in the atmosphere (Watson and others, 1990). There are uncertainties in accounting for all the CO_2 emissions which do not remain in the atmosphere. Other potential long term carbon sinks (Tans and others, 1990) have not been quantified but would shorten the effective residence time for CO_2 .

The contribution to the greenhouse effect of CH_4 relative to CO_2 is important when considering the merits of fuel substitution from coal to natural gas or a carbon tax. The global warming potential of one mole of CH_4 relative to that of CO_2 ranges from 20–35 or more depending on the assumptions of different authors (Selzer, 1989; Zittel and Selzer, 1990). This uncertainty is compounded by differing values for the effective residence time of CO_2 , resulting in a wide variety of numbers for the relative global warming potential of CH_4 : CO_2 (see Table 1). The estimates in Table 1 are ranked from the simplest (no consideration of the differences in residence times for CO_2 and CH_4 and a molar global warming potential of 27) to the most sophisticated by Lashof and Ahuja (1990) and Shine and others (1990). The latter use oceanic models for estimating the residence time of CO_2 , more complex chemistry for the indirect effects and include the concept of integrating over the short or long term. In the long term, CO_2 assumes greater importance relative to CH_4 than in the short term. Despite current limitations, the concept of applying global warming potentials over different time horizons is the best available means of evaluating emission abatement scenarios but requires further validation.

The contribution of global coal use to the enhanced greenhouse effect is evaluated from CO_2 emissions in 1988 for the low and high estimates of the contribution from deforestation and land use adopted by the IPCC (see Table 2). Shine and others (1990) assume 7.1 GtC/y for the total CO_2 emissions from human activities in 1990. This lies within the range for total CO_2 shown in Table 2 but implies a low value for deforestation and land use if the 1988 CO_2 emissions of 5.7 GtC/y for fossil fuel are extrapolated to 1990. There is little difference between the contributions for one year or over 100 y. Coal, oil and gas would contribute about 20%, 20% and 8% respectively. There would be an additional 1% and 3% for CH_4 from hard coal mining and gas pipeline leakage respectively, assuming these sources contributed 6.3% and 20.8% of CH_4 emissions from human activities (World Resources Institute, 1990). There would also be small contributions from N_2O and NO_x emissions but these have not been quantified reliably.

Average CO_2 emission factors for bituminous coal, crude oil and natural gas, based on the higher heating value (HHV), are 24.1, 19.9 and 13.8 gC/MJ (Marland, 1983; Marland and Rotty, 1984). Those for the brown coals assessed by Couch (1988) averaged 25.2 gC/MJ. Anthracites have CO_2 emission factors of about 26 gC/MJ. When allowance is made for the effect of CH_4 , in terms of its CO_2 equivalent, the emission factor for natural gas increases relative to that of coal and oil due to CH_4 leakage from natural gas distribution systems. Mitchell and Sweet (1990) applied the concept of global warming potentials and estimated that coal and natural gas would have the same emission factors at a CH_4 leakage rate of 11.5% over a 100 y time horizon. Over 20 y, the breakeven point would occur at a CH_4 leakage rate of about 5%. Such CH_4 leakage rates are not uncommon in some systems. However, it is unlikely that leakage from new gas supplies for large scale use would exceed 1% (James, 1990).

Similar evaluations including hard and brown coals in the FRG (Zittel and Selzer, 1990) conclude that, over short time horizons, brown coal has a lower effective CO_2 emission factor than hard coal if most of the CH_4 from hard coal mining is vented to the atmosphere. The two fuels would be equal over the 100 y time horizon.

Energy consumption or emission penalties are incurred when coal is upgraded. For example, a coke from a bituminous coal with a CO_2 emission factor of 24.1 gC/MJ (HHV) had an effective emission factor of 31.8 gC/MJ when coal production overheads were included (Thurlow, 1990). Synthetic fuels

from coal generally have much higher CO₂ emission factors than the original coal - even where gasoline is produced at a high thermal efficiency of 65% or more, the CO₂ emission factor is 38 gC/MJ. However this penalty will be offset by the lower CO₂ emission factor and improved combustion efficiency of the resulting synthetic fuels.

ABATEMENT

The proportion of coal used in power generation is about 73% in OECD countries (IEA, 1990) but worldwide it is probably around 50% (Matthews and Gregory, 1989). This section will therefore concentrate on technologies which decrease CO₂ emissions per unit of useful energy in the large scale use of coal for power generation.

The efficiency of coal use in steam power plants has already improved greatly this century. For example, in the FRG increasing operating pressures and temperatures and the use of steam reheat in conventional power plants have resulted in a reduction in the heat rate from 35 MJ/kWh in the early 1900s to about 9 MJ/kWh in the 1980s (Schilling, 1990). Emissions and waste heat losses are now only about 25% of what they were in the early steam based power plants.

Current trends show that further improvements are achievable, both on conventional plants and using more advanced technologies (see Table 3). Hence the use of supercritical steam conditions (Riedle and others, 1990) should reduce CO₂ emissions by 20% compared to a conventional 500 MWe pulverised coal (PC) plant with wet limestone flue gas desulphurisation (FGD) and sub-critical steam conditions (US DOE, 1990). Co-firing with natural gas or oil also results in reductions in CO₂ emissions due to a lower emission factor of the blending fuels. The use of flue gas recycling (Wolsky and Brooks, 1989) or selective H₂ combustion (Steinberg and Grohse, 1990) appears to offer 100% reductions of CO₂ emissions but at considerable energy costs for the separation and disposal of CO₂ or carbon.

The use of combined cycles with gasification, pressurised pulverised coal or fluidised bed combustion (PFBC) should achieve CO₂ reductions of 13-26%. At the upper end of this range are the hybrid PFBC gasification topping cycle developed by British Coal (Dawes and others, 1990) and a new scheme for pressurised pulverised coal combustion (Weinzierl, 1990). In the latter, pulverised coal is burnt under pressure in an ash slagging combustor with gas expansion through a gas turbine. Pulverised coal-natural gas combined cycles with supercritical steam conditions are expected to achieve a reduction in CO₂ emissions of about 37% (Hebel and Kotschenreuther, 1990).

Commercial MHD generators could reduce CO₂ emissions by up to 27% in the near term and even more in future with a greater efficiency of heat recovery (Morrison, 1988). The development of molten carbonate and solid oxide fuel cell technology is currently in its infancy but is potentially capable of reducing CO₂ emissions by up to 44% when used with gas/steam turbine combined cycles (Kinoshita and others, 1988).

Finally, where waste heat can be used in cogeneration or combined heat and power such as in Denmark (Mortensen, 1989), CO₂ reductions approaching 60% are achievable even with conventional coal combustion technologies.

CONTROL TECHNOLOGIES

In contrast to abatement measures, the control of CO₂ emissions by recovery and disposal is generally regarded as a last resort owing to the high energy costs involved. A recent study by the US Electric Power Research Institute (Smelser and Booras, 1990) concluded that 90% CO₂ recovery by MEA scrubbing, solvent regeneration, compression and removal for disposal in the ocean at a depth of 457 m would incur an energy penalty of 370-374 kWh/t CO₂. The busbar cost of electricity would increase by 159-178% relative to a coal-fired power plant without CO₂ control. These results lie within a range of 222-988 kWh/t CO₂ for the energy penalty estimated by several authors for control technologies using absorption or cryogenic separation at 90% CO₂ removal or membrane separation at 80% removal. The estimates of energy use are in the range of 21-95% of the energy released from coal combustion (Smith and others, 1991).

Disposal options include the oceans or sites on land such as salt domes and depleted natural gas or oil

fields. For disposal in the oceans, the transfer of CO_2 as solid blocks of dry ice or hydrate was found to be less practical and more costly than gas or liquid injection owing to the tonnage involved. It is desirable to inject the CO_2 into the ocean at depths $>1,000$ m and preferably at 3–4,000 m in order to minimise the transfer of CO_2 from the ocean to the atmosphere (Hoffert and others, 1979). More research is required on the long term effects of macromixing in the oceans on CO_2 retention in the deep ocean. On land there is potential for storage of liquid CO_2 in salt domes. For example in the USA, the existing storage capacity of strategic crude oil reserves could accommodate about 15 GtC of CO_2 emissions. Global 1980 natural gas production values suggested that the storage capacity of depleted gas reservoirs could increase at about 0.7 GtC/y with an overall capacity as large as that of past and present reserves of natural gas (Baes and others, 1980). Storage of CO_2 in depleted oil fields can be combined with its use as a solvent for enhanced oil recovery by miscible flooding. In the USA it is estimated that this use could only take up 7.7 MtC/y of CO_2 (Abel and others, 1989). However, there would be a substantial release or recycling of injected gas at active production wells.

Alternative means of controlling CO_2 emissions include its use or recycle by natural or industrial processes. A recent study by Martin and others (1990) of essential growth nutrients in the Antarctic ocean shows that the correction of an iron deficiency could promote a thirty fold increase in phytoplankton growth in the outer continental sea. Estimates indicate that increased growth by iron seeding could sequester up to 1.6 Gt C/y of CO_2 over a 50 y period but such an action is undesirable because of unknown ecological effects (Baum, 1990). On land, commercially managed forests in temperate ecosystems could sequester 2.9–5.4 Gt C/y but securing the necessary land areas of 465–733 Mha (Marland, 1989; Sedjo and Solomon, 1989) would pose problems. Unused land in tropical ecosystems (865 Mha) that previously supported forests might however be used to accumulate up to 1.5 Gt C/y over a 100 y period (Houghton, 1990). Two tropical reforestation projects by US and Dutch utilities are to offset the CO_2 emissions from a new 180 MWe coal-fired power plant in the USA and two 600 MWe coal-fired power plants in the Netherlands (Flavin, 1989; EER, 1990). The reuse of CO_2 in fuel or chemical synthesis is economically viable only if the total energy used can be produced at a lower cost than the fuel or chemical value of the product. Here, biofuels, relying on solar radiation as a 'free' energy source for direct removal of CO_2 from the atmosphere, offer a means to slow the growth of fossil fuel use whilst simultaneously reducing the net accumulation of CO_2 in the atmosphere. The present consumption of CO_2 for process use and the synthesis of industrial chemicals suggests that this sector is unlikely to make a significant contribution to reducing the overall global emissions of CO_2 .

CONCLUSIONS

The contributions of the greenhouse gas emissions to the enhanced greenhouse effect are expressed in terms of the equivalent global warming potential of CO_2 when policies to reduce emissions are evaluated. This exercise is however complicated by indirect effects from some, as yet, unquantifiable chemical reactions and by uncertainties regarding the effective residence time of CO_2 in the atmosphere. Thus the concept of applying global warming potentials of greenhouse gases over different time horizons is the best available means but requires further revision and evaluation. It is clear that CO_2 is the most important greenhouse gas from coal. However, consideration of CH_4 affects the relative ranking of different fossil fuels as greenhouse gas emitters. Hence high CH_4 leakage rates of 5–10%, such as occur in some natural gas distribution systems, would virtually offset the advantage of the lower CO_2 emission factor for natural gas over coal; although the leakage rates of $<1\%$ claimed for new gas distribution systems would make little difference. Coal use worldwide is responsible for about 20% of the enhanced greenhouse effect, half of which is attributable to coal-fired power generation.

There are many options for the abatement of emissions from coal use in power generation. Apart from reducing the overall demand for energy by conservation, there are technologies which will reduce emissions from an improved efficiency of coal use. Existing technologies based on supercritical steam cycles can achieve CO_2 emission reductions of up to 20%. Over the next decade, technologies such as PFBC and gasification combined cycle offer improvements up to 25% or 37% with the use of pulverised coal-natural gas combined cycles. For the future, MHD and fuel cells should reduce CO_2 emissions by 17–44%. All of these technologies could be improved further with co-generation for CO_2 emission reductions approaching 60% – close to that required to stabilise concentrations of CO_2 in the atmosphere.

The control of CO₂ emissions by recovery and disposal appears to incur too high an energy penalty with uncertain environmental consequences for a practical solution. However systems which offer the possibility of the combined removal of CO₂, SO_x and NO_x might have some role in future in conjunction with energy savings. Reafforestation and recycling of CO₂ in fuels potentially have more scope for reducing the concentration of CO₂ in the atmosphere but probably not on the scale required to stabilise it.

Considerable progress has been made to improve efficiency for economic reasons in the past. Now there is an added impetus to achieve more for both economic and environmental benefits. There is scope for reducing emissions from coal use by both abatement and control measures but their success depends on how quickly they can be implemented.

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Table 1 Summary of estimates of CH₄: CO₂ global warming potential

Global warming potential CH ₄ : CO ₂ (molar)	Effective residence time, y		Allowing for indirect effects	Reference
	CO ₂	CH ₄		
27	(10)	10		
3.5	100	10	CO ₂	(Selzer, 1989)
6	60	10	yes	(Pearman, 1989)
10	120	10	O ₃ , CO ₂	(Rohde, 1990)
3.7	230	14.4	O ₃ , H ₂ O, CO = CO ₂	(Lashof and Ahuja, 1990)
12 (1% discount of future)	45			
17 (20 y)*	120	10	O ₃ , H ₂ O, CO, NO _x , NMHC	(Shine and others, 1990)
5.7 (100 y)*				
2.5 (500 y)*				

* integration time horizons

NMHC = non-methane hydrocarbons

Table 2 Contributions to the greenhouse effect (Boden and others, 1990; Shine and others, 1990; Watson and others, 1990)

	CO ₂ emissions, 1988		Greenhouse effect, %	
	Gt C/y	%	over 1 y	over 100 y
Coal	2.4	37-28	21-16	23-17
Oil	2.4	37-28	21-16	23-17
Gas	0.9	14-11	8-6	9-7
Fossil fuel	5.7	88-68	50-38	54-41
Cement manufacture	0.2	2.3-1.8	1.3-1.0	1.4-1.1
Deforestation and land use	0.6-2.6	9-31	5-17	6-19
Total CO ₂	6.5-8.5	100	56	61

Table 3 Summary of CO₂ abatement measures for power generation (Smith and others, 1991)

Technology	Steam conditions	Net power plant efficiency (LHV), %	CO ₂ emissions, gC/kWh	CO ₂ reduction, %	Comments
Conventional					
PC + FGD	sub-critical	35	262.1	0	reference plant for CO ₂ emissions
PFBC	sub-critical	37	257	1.95	in-bed desulphurisation, demo _x
PC + FGD, demo _x	super-critical (250 bar)	41.2	228.1	13	540-560°C, 0.065 bar condenser
PC + FGD, demo _x	super-critical (300 bar)	45	209.4	20	600°C, 0.03 bar condenser
PC-natural gas cofiring	sub-critical	36-37.5	233.3-242.1	7.4-11	15% natural gas input
PC design - coal oil mixture	sub-critical	35	251.2	4.2	40% fuel oil input
PC-CO ₂ flue recycle	sub-critical	27.6-28.6	325.9-337.5	(100)	additional energy use for CO ₂ gas disposal
PC design, H ₂ combustion	sub-critical	8.4	-	(100)	energy use for H ₂ separation 76% of coal heating value, additional energy use for carbon disposal
Combined cycles					
IGCC	sub-critical	43	212.7	18.9	low temperature gas cleaning
PFBC	sub-critical	42	226.7	13.5	high temperature gas cleaning
PFBC-gasification cycle	super-critical (250 bar)	48-49	195.6-199.5	23.9-25.4	high temperature gas cleaning
Pressurised pulverised coal	sub-critical	48	194.2	26	additional energy used for gas cleaning, CO ₂ emissions from desulphurisation
PC-natural gas + FGD, demo _x	sub-critical	40	202.1	22.1	33% natural gas input
PC-natural gas + FGD, demo _x	super-critical (300 bar)	49	166.1	36.6	33% natural gas input
MHD	sub-critical	40-45*	192.5-216.5	17-27	additional CO ₂ emissions from desulphurisation
Fuel cells	sub-critical	45-58*	147.1-192.5	26-44	additional CO ₂ emissions from desulphurisation
Cogeneration					
Combined heat and power (CHP)	sub-critical	77-84	108.9-116.5	54-59	range for PC + FGD, PFBC, and PFBC-gasification cycle

* HHV

DEMONSTRATION PLANTS: ARE THEY NEEDED?

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INTRODUCTION

In the evolution of new technologies that demonstrate and challenge existing practice, the concept of a demonstration plant is used to bring the process of research, development and demonstration to conclusion. The incorporation of a demonstration plant in the strategy of realizing commercial application of a new technology was a new step introduced by the availability of support from federal agencies that provided the necessary funds required for large plant scale-up and sustained operation.

A. Commercial Plant Following Bench Scale or Pilot Plant Tests

In the chemical industry commercial plants are often constructed based on small scale (less than 1 ton/day) continuous pilot plant trials where there is a large incentive to realize commercial production of a product that is produced at lower cost or that has improved quality over competing routes. Often the commercial plant is built based on kinetic and yield information from limited pilot plant data (10-50 lbs/day).

In these instances homogeneous phase systems are scaled up to commercial sized plants. Engineering usually does not represent a significant change from existing practice that is in existence. Examples of new technology usually include improvements in reactor design (contacting) and/or in catalyst formulation, representing the primary new steps being proven or modified. Adjustments and plant modifications are made at the time of commercial plant "breakin" and startup. An extra allowance is provided for the "first of a kind" commercial plant which usually is 5-25% of the capital cost of the plant. The advantage of this approach is rapid commercial application that takes advantage of an economic incentive that is significantly large.

B. Modification of An Existing Commercial Plant

In some cases an existing commercial plant can be modified to incorporate new features of a technology. For example, an improved high capacity gasifier may easily be incorporated into an existing coal gasification

commercial plant. This strategy is often used to avoid the cost of installing the raw materials receiving, feed preparation, and product recovery steps which may be relatively unchanged and that may need no modification. Thus, a new technology such as a high capacity reactor can be demonstrated at relatively modest cost at an existing commercial plant. The new gasifier may represent 10-15% of the total capital requirements.

Projects that are demonstrating sorbent absorption of sulfur dioxide in the hot gas path of an existing power plant are examples of this approach. In the petroleum industry new more selective or aging resistant catalysts are incorporated into an existing plant. This approach is usually a low cost approach to bring a new technological advance to commercial realization.

C. Utilization of Large Pilot Plant

For the situation where there is a change in phase a step wise series of continuous bench scale and pilot plants are used to:

- (1) provide scientific information on kinetics, selectivity, product characteristics, yields, etc. Often this information is empirical. The chemistry and mechanisms are not very well understood.
- (2) provide engineering data on equipment size requirements, maintenance, materials information, engineering design and safety data, and design of special equipment and subsystems. Catalyst life data are defined based on sustained "steady state" operations performed.

On the basis of the continuous pilot plant trials at increased capacity scale up uncertainties are also defined. In the largest pilot plants operated the smallest commercial size equipment from vendors are used to minimize risk of equipment scale up at a pioneer commercial plant. Table 1 shows the features of a pioneer commercial plant. It should be noted that an integrated program requires various size pilot plants to be operated simultaneously. Thus, the program often requires small and large pilot plants to be operated on feed materials used in a commercial plant. The objective of the integrated program is to obtain the engineering equipment design data and yield and product quality information that permits the pioneer plant to be constructed with ordinary financial risk. Trained operations personnel from the large pilot plant serve as a cadre of experience specialists for the pioneer plant.

D. Demonstration Plant

In the development of new technology in which solid feeds (coal or shale) are converted the concept of a Demonstration Plant has been introduced. It usually represents a single full scale train (1000-5000 Tons/day) of a

TABLE 1.
Pioneer Plant Objectives

- Commercial pretested equipment used to prove the technology in full scale module.
- Establish firm basis for investment, operating procedures, costs of commercial plants.
- Establish permitting procedure for plant, transportation of product, utilization by customers
- Complete testing, and handling procedures and health effects in markets
- Ability to expand pioneer plant to full sized commercial plant.

commercial plant that represents an engineering proof of the information from large pilot plants (100-500 tons/day). An important aspect of such plants is the culmination as a technical spectacular.

Such spectacles, which challenge existing practice, represent step outs that are important "first of a kind" culminations concluding research that may have taken 10 years or more to accomplish. Being first is always an exciting challenge for scientists and engineers.

A demonstration plant has many purposes and objectives including the challenge of using expert knowledge and organizing and managing the skills necessary to achieve technical innovations. Recent demonstration plant experience that featured solid feeds to (coal and shale) indicates that a minimum of 3 years and more appropriately 5 years or more of operations are required to obtain robust information on maintainability and reliability from a utility industry perspective.

Table 2 shows the features of a demonstration plant and its objectives. For a conservative industry like the electric power industry, that must be prudent and is not used to accepting significant financial risk, the added comfort of robust cost, design, reliability and maintenance practice information is an important aspect favoring demonstration plants that represent technological innovations for generating electricity.

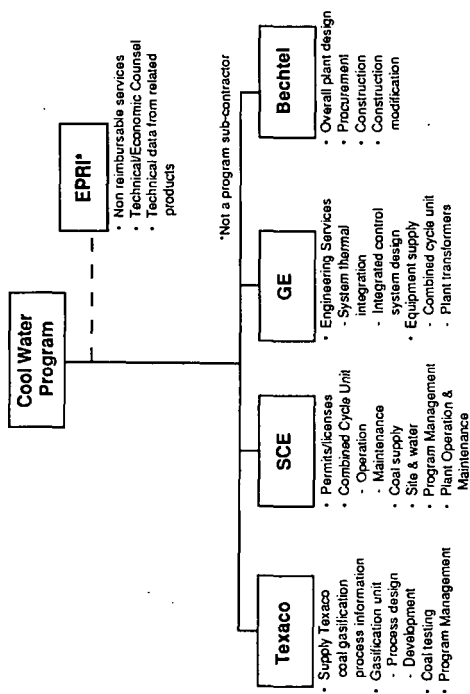


FIGURE 1. Responsibilities of Program Participants

TABLE 2.

Objectives of Demonstration Plant

- Save time to commercial production of products, introduction of new technology
- Complete engineering development from small equipment
- Confirm scaleup parameters, establish sources of commercial equipment
- Operate integrated flow sheet, obtain operating experience, confirm design
- Confirm economics of commercial plant and expand demonstration plant if warranted
- Produce large samples of products for test by customers

While the cost for a demonstration plant is high, new technology acceptance will depend on their outcome. The learnings from demonstration plants guarantee efficient smooth start up and operation of subsequent commercial plants that can reliably proceed with low financial risk.

While there are many advantages for a demonstration plant a major shortcoming is its cost. In the case of the Cool Water program about \$300 million was required to build and achieve reliable operations over the course of the 5 year operating program.

Figure 1 shows the responsibilities that were defined for each of the major participants that supplied funds for the Cool Water Program. At the conclusion of the program the demonstration plant was shut down. It is imperative to carefully define the scope of each organizations responsibility at the programs inception.

E. The Cool Water Program

Figure 2 shows a block diagram of the Cool Water plant.

Cool Water is a nominal 120-MW IGCC power plant that uses the Texaco Coal Gasification Process to produce a medium-Btu fuel gas. The gas is combusted in the combined cycle portion of the plant to produce electricity. The plant is designed to process up to 1000 tons of coal per day mixed with water, however, rates as high as 1200 tons per day were achieved. The coal-water slurry is gasified with oxygen at a pressure of approximately 600 psig and a temperature of between 2100 °F and 2500 °F. The raw syngas from the gasifier is first cooled in radiant and convective coolers that generates steam for use in

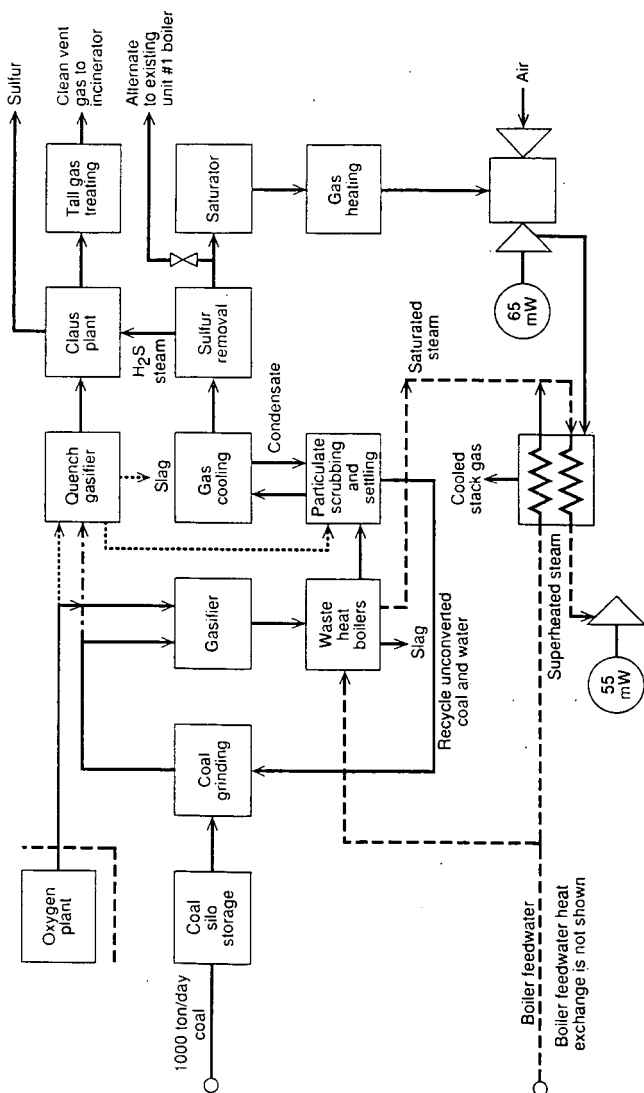


FIGURE 2. Block Flow Diagram of the Cool Water Coal Gasification Plant

power production. The slag solidifies during its fall through the radiant cooler and collects in a water sump at the bottom of the cooler, where it is removed periodically using a lockhopper system.

After leaving the radiant and convective syngas exchangers, the cooled synthesis gas enters a water scrubber where any entrained particulate is removed. After the scrubber, low level heat is recovered from the gas in a series of exchangers, and then air and cooling water trim coolers reduce the gas temperature to approximately 100 °F. Next, sulfur is removed, primarily in the form of hydrogen sulfide, in a Selexol absorber. A sulfur-rich acid gas is stripped from the Selexol solution and routed through a SCOT/Claus sulfur recovery unit to produce an elemental sulfur product. Following sulfur removal, water is added to the desulfurized gas in a syngas saturator to suppress NO_x formation during combustion.

Subsequently the synthesis gas is fired in a GE MS7001E gas turbine to produce electricity. The exhaust gas from the combustion turbine passes through a Heat Recovery Steam Generator (HRSG) where saturated steam formed in the syngas coolers, and in the middle section of the HRSG, is superheated. After superheating, the steam is utilized in a steam turbine for additional electricity production.

F. Definition of Incentives

The Coolwater Program was started in the late 1970's. A major incentive was the control of emissions of an IGCC plant compared to a pulverized coal plant as shown in figure 3. Demonstration of these environmental features was considered an important incentive for the project.

In addition economic analyses showed that electricity from IGCC plants would be cost competitive with conventional coal plants and offer higher efficiency routes to electric power.

At the time of the start of the program oil and gas prices were expected to escalate in real terms. By the last year of the operating period economic dispatch of the facility on the Southern California Edison Co. system was projected. Projections of oil prices of \$40 a barrel or higher were expected.

At the conclusion of the program the plant was not competitive with low cost natural gas in conventional generation plants that produce electric power at a cost of about 3-4¢/Kw-hr, or about 50% of the cost of electricity from the Cool Water plant.

Environmental Trade-offs for Coal-Based Technologies

Flue gas desulfurization (FGD) units and fluidized-bed combustion—both atmospheric (AFBC) and pressurized (PFBC)—have been effective in reducing emissions of SO_2 and NO_x from coal-fired plants, but at the cost of producing substantial volumes of solid waste. Gasification-based generation options (IGCC, IGHT, and IGMCF) have the potential to cut those airborne emissions further and minimize solid waste without imposing an efficiency penalty on the overall system.



FIGURE 3. Environmental Trade-offs for Coal-Based Technologies

Table 3 shows a summary of the results from the Cool Water program which represented the "know how" produced

TABLE 3.
Major Information From
Demonstration Plants

Management

- Firm basis for capital and operating costs
- Firm basis for plant performance as base load or load following
- Definition of management structure
- Plant reliability as a variable for design
- System integration and performance
- Environmental data for commercial plant licensing

Technical

- Design and equipment requirements
- Performance of equipment from sustained operations on a variety of coals
- Comprehensive information on materials of construction, components, subsystems
- Comprehensive details on environmental impacts and equipment alternatives
- Health and safety information
- Staffing requirements
- Training of future users of the technology, simulator

CONCLUSIONS

Currently a large number of demonstration projects are being performed in the U.S. with funding from federal and private sponsors. The Cool Water Program was a technical success in that all of its objectives were met or exceeded during the 5 year operating period. It's major achievement was to launch a new technological option for the world wide electricity industry--the IGCC plant- which is a clean use of coal.

Of prime importance is agreement by the sponsors on its, objectives and participation by organizations who take the information forward to commercial projects that follow.